

Remote Measurements of
Air Pollutants

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REMOTE MEASUREMENTS OF AIR POLLUTANTS

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ABSTRACT

There continues to be a great need for the development of remote measurement systems for air pollution and other applications. Accordingly, the objectives of this work were (1) the development of remote measurement systems, (2) the demonstration of their capabilities, and (3) the development of new laser sources for remote sensing applications. This research on the remote measurement of air pollutants has been conducted jointly by SRI International and Stanford University.

This research focused on the differential absorption laser radar or DIAL technique. The DIAL technique has been demonstrated to offer much better sensitivity and longer range for detection of trace constituents of the atmosphere than the competing Raman and fluorescence techniques. Demonstrations of remote measurements have been made of NO_2 in the visible spectral region; SO_2 and O_3 in the ultraviolet; HCl , CH_4 , SO_2 , and N_2O in the mid-infrared; H_2O , ethylene, and temperature in the far-infrared; and CH_4 in the near-infrared. Results of this research indicate that remote measurements at high sensitivity and long range are feasible using currently available technology.

The applications of this research encompass measurement of the environment, exploration of new energy resources, and military uses. Specific applications of DIAL systems include: enforcement of air pollution regulations, testing of air pollution models, real-time investigation of the atmospheric chemistry of air pollutants, petroleum exploration via detection of light hydrocarbons indicative of subsurface deposits, detection of pipeline leaks, detection of gases of military interest, and exploration of regions of geothermal activity.

The early pioneering measurements performed under this grant in the visible and ultraviolet spectral regions have already led to the design, construction, and use in field operations of two generations of ground-mobile systems for measurement of NO_2 , SO_2 , and O_3 . This research has

also led to development of remote sensing capabilities in the ultraviolet to measure biological aerosols and at far-infrared wavelengths to measure gases of military interest. Pioneering measurements in the mid-infrared have led to additional research and development on the utilization of DIAL systems for petroleum exploration. Other spin-offs from research under this grant include studies of remote measurement capabilities of DIAL systems mounted in spacecraft, aircraft, and ground-mobile systems.

Because of the many varied applications of research performed under this grant, the results of this research will continue to impact many different areas of the scientific applications community.

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I INTRODUCTION

A. Objective

The primary objective of this research was to investigate laser techniques for performing remote measurements of air pollutants. Point monitors have a capability limited to the measurement of the concentration of a pollutant at a single location. The additional capability of a remote measurement system has extensive applications in air pollution enforcement, monitoring of ambient air quality, testing of plume models, investigations of atmospheric chemistry, and in other areas of research on air pollution.

B. Background

Remote monitoring of the atmosphere with laser sources has now been actively pursued for over a decade. However, only recently has the development of tunable lasers with adequate energy and spectral purity reached a state that meets remote monitoring requirements. Thus, in the past few years tunable lasers have allowed an expanding range of applications and measurements to be made by remote monitoring methods.

The laser as a source offers four unique features unavailable in incoherent light sources: high power, spatial collimation, spectral purity, and coherence. Remote monitoring uses all of these laser properties in the measurement. In addition to being generally useful, the laser transmitter must also be tunable so that atomic or molecular absorptions can be probed to yield information about species concentration.

The three primary remote measurement techniques using lasers are: the differential absorption lidar or DIAL technique, fluorescence, and Raman lidar. The DIAL technique has been established as the technique with the highest sensitivity and longest range for measurements of small amounts of gases in the troposphere. The magnitude of the signal received from the three techniques is related to sensitivity and range of the

system. The superiority of DIAL is based on the much larger scattering cross section.

DIAL systems transmit pulses of narrow bandwidth laser radiation into the atmosphere. Backscattering is obtained from naturally occurring aerosols. The laser is tuned to two wavelengths: one wavelength is absorbed by the gas of interest, and the other is not. The magnitude of the return signal on the nonabsorbed line is used to effectively calibrate the gain of the optical and electronic subsystems, and the backscatter from the naturally occurring aerosols. The difference between the absorbed and nonabsorbed lines is a measure of the concentration of the absorbing species. Pulsed operation yields range-resolved measurements similar to radar operation. Single-ended path-averaged measurements can be obtained with the same system by pointing the lidar at a topographic target and by observing the return signals.

The DIAL technique has been pursued jointly by SRI International and Stanford University on a grant from the National Science Foundation (NSF). SRI has investigated the use of continuously-tunable dye lasers emitting ultraviolet and visible wavelengths and discretely-tuned infrared lasers for remote measurements of pollutants. Stanford University has developed the continuously-tunable optical parametric oscillator (OPO) as a laser source and has applied it to remote measurements.

This successful joint effort has led to pioneering demonstration measurements of DIAL on many gases in the ultraviolet, visible, near infrared, mid-infrared, and far-infrared spectral regions. A large number of publications and presentations have documented and widely disseminated the results of this research. The earlier work on this grant using ultraviolet and visible wavelengths has already led to the construction and use in the field of a self-contained mobile lidar system. A second and improved version of this system is currently under construction at SRI for the Electric Power Research Institute (EPRI). Results of the more recent research under this grant in the infrared spectral regions are expected to lead to field hardware in the near future.

II RESEARCH RESULTS

A. SRI International

1. Measurements of NO₂ at Visible Wavelengths

The first undertaking at SRI on the grant from NSF was investigation of the remote monitoring capabilities of tunable dye lasers. A demonstration measurement of remote sensing of NO₂ was performed using the system shown in Figure 1. This work was reported by Grant et al. (1974). The dye laser was tuned to several wavelengths near 440 nm. A sample cell was located at a range of 365 m and known concentrations of NO₂ were measured remotely using the lidar. Backscattered energy both in front of and behind the chamber was used to infer the concentration of NO₂. The backscattered signal was collected using a telescope and focused on a photomultiplier tube. The output signal was digitized, stored on magnetic tape, and displayed in real-time. The experimental parameters are shown in Table 1.

A summary of the comparison of these first lidar measurements of NO₂ with measurements made with an in situ monitor are shown in Figure 2. The general agreement of the slope of the experimental data with the line of expected values indicates that the lidar is functioning correctly and the absorption coefficient required for the data reduction is accurate. The measured uncertainty for a single pair of pulses was 50 ppb-km. It was established that, by using an improved laser and digitizer, a sensitivity of 5 ppb-km could be achieved with a single pair of pulses. Much later we demonstrated that this factor of 10 improvement was in fact achievable in practice. This is discussed in more detail later in this report. Multipulse integration will improve the sensitivity even further as the square root of the number of pulse pairs.

Local ambient concentrations of NO₂ are usually between 10 ppb and 100 ppb. Therefore, useful remote measurements at ambient concentrations have been demonstrated with the tunable dye laser system.

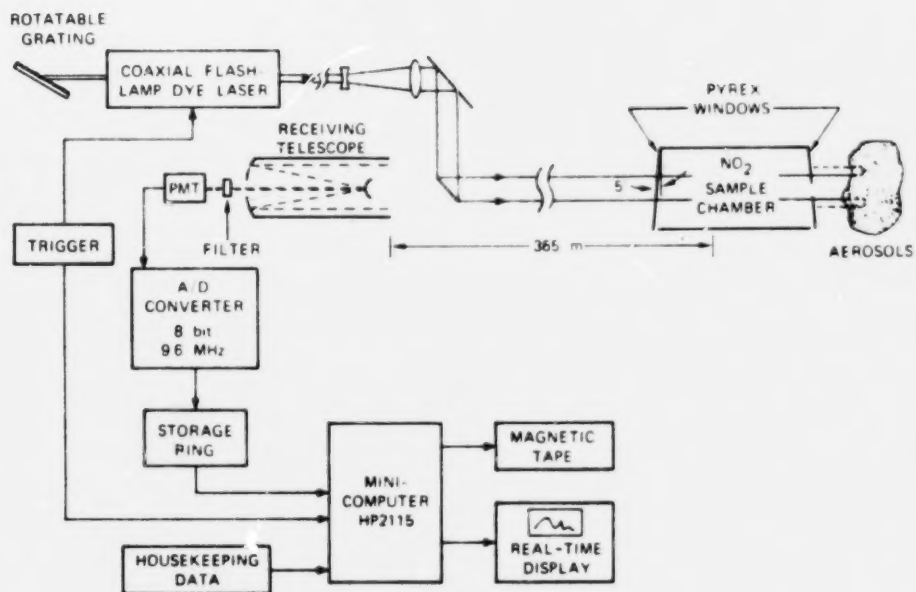


FIGURE 1 APPARATUS USED TO MAKE CALIBRATED, REMOTE MEASUREMENTS OF NO_2

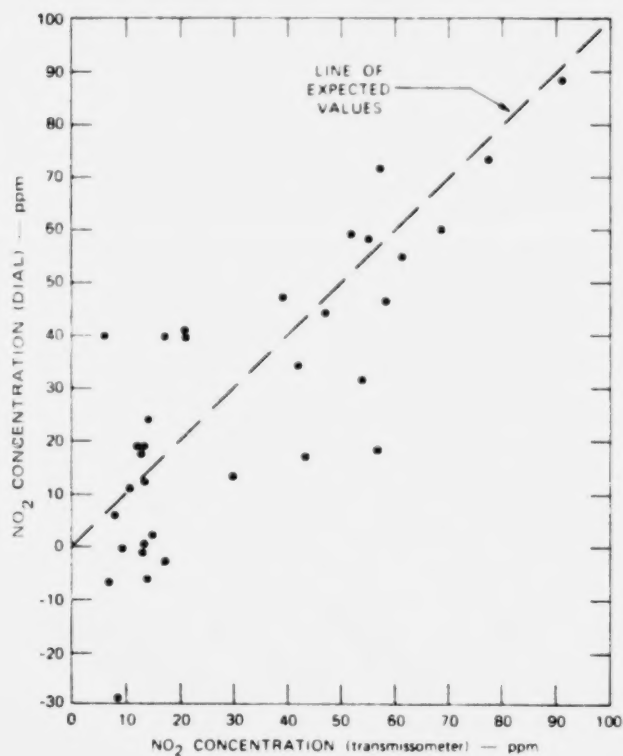


FIGURE 2 VALUES OF NO_2 IN THE SAMPLE CHAMBER AS MEASURED BY DIAL AND COMPARED WITH TRANSMISSOMETER MEASUREMENTS

Table 1

EXPERIMENTAL PARAMETERS FOR MEASUREMENT OF NO₂

Laser	
Transmitted energy	4-8 mJ
Pulse length	250 ns (3 dB)
Transmitted beam divergence	0.3 mrad
Dye	4,6-dimethyl-7-ethylamino-coumarin (2 X 10 ⁻⁴ M/l in methanol)
Receiver	
Aperture	0.028 m ²
Filter	4400 Å, 160 Å (3dB), 57% peak transmission
PMT	ITT FW-130
Transmissometer	
Lamp	Tungsten filament
Filter	Corning glass (No. 3-73 and No. 5-58)
PMT	RCA 1P28
NO ₂ Spectral Data	
Wavelength (Å)	Absorption cross section (cm ⁺²)
4418	3.76 X 10 ⁻¹⁹
4448	6.33 X 10 ⁻¹⁹
4465	3.83 X 10 ⁻¹⁹
4481	6.70 X 10 ⁻¹⁹
Atmospheric Conditions	
Time	9-12 p.m.
Visibility	16-24 km
Ambient NO ₂	10-30 ppb

2. Measurements of SO_2 and O_3 at Ultraviolet Wavelengths

No other important pollutants absorb visible radiation, so we turned our attention to the ultraviolet spectral region. SO_2 and O_3 can be measured using frequency-doubled dye lasers operating near 300 nm.

The experimental apparatus shown in Figure 3 was used to measure SO_2 and O_3 concentrations in a sample chamber located 306 m away. This work was reported by Grant and Hake (1975). The primary difference between this apparatus and that used to measure NO_2 is that an ADA crystal is used to frequency double the output radiation. The experimental parameters are shown in Table 2.

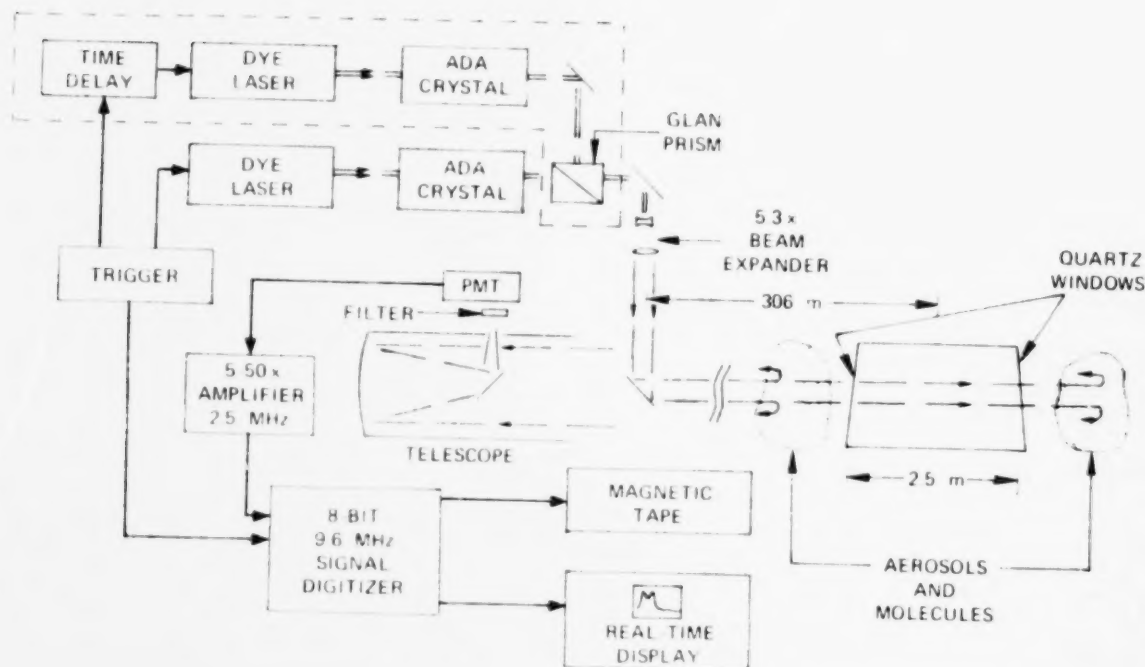


FIGURE 3 BLOCK DIAGRAM OF THE EXPERIMENTAL APPARATUS USED TO MEASURE SO_2 AND O_3 . The equipment enclosed in the dashed lines was not used in these measurements but could be added to provide nearly simultaneous transmission at two wavelengths.

Table 2

EXPERIMENTAL PARAMETERS FOR MEASUREMENT OF SO_2 AND O_3

Laser	
Energy	≈ 15 mJ
Dye	Rhodamine 6G (3.5×10^{-2} M/l in MeOH)
Beam diameter	≈ 2 mm (3dB)
Beam divergence (far field)	0.7 mrad (3 dB)
Pulse length	250 ns (3 dB)
Grating	316 l/mm, used in ninth order
Linewidth	≈ 1 Å
Repetition rate	0.017 Hz
Second-Harmonic-Generating Crystal	
Type	Ammonium dihydrogen arsenate
Length	2 cm
Index matching fluide	Fluorocarbon 77
Conversion efficiency for 15-mJ funda- mental	5%
Pulse length	≈ 200 ns (3 dB)
Receiver	
Aperture	0.056 m ²
Filter	293 nm, 13-nm half-width
PMT	27% peak transmission
Optical efficiency from ADA crystal through PMT	RCA 7200
	0.01
Transmissometer	
Lamp	Deuterium
	285.7 nm, 3-nm half-width
Filter	17% peak transmission
PMT	RCA 1P 28
Accuracy	± 7 ppm
Spectral Data	
Wavelength (nm)	Absorption Coefficient ($\text{cm}^{-1}\text{atm}^{-1}$) (base e)
	SO_2 O_3
292.3	26 28
293.3	14 22
294.0	
Atmospheric Conditions	
Time	19 Dec. 1974, midnight to 5:00 a.m.
Visibility	10-20 km
Molecular scattering coefficient (calcu- lated)	$\approx 0.14 \text{ km}^{-1}\text{atm}^{-1}$
Aerosol scattering (extinction) coeffi- cient (observed)	$\approx 0.5-0.7 \text{ km}^{-1}$
Ambient SO_2 , NO_2 , and O_3 concentrations	< 0.03 ppm

The results of the measurements of SO_2 are shown in Figure 4. These results show that the lidar agrees well with the point monitor in measuring the increasing and then decreasing concentrations of SO_2 . The uncertainty in this measurement was 60 ppb-km.

The results of the measurements of O_3 are shown in Figure 5. Again, the lidar results agree with the point monitor, and in this case the uncertainty was 120 ppb-km. The larger uncertainty in the measurement of O_3 is caused by the small differential absorption coefficient at the wavelengths used for this measurement. The use of more advantageous wavelengths, an improved laser, and better electronics will yield a factor of 10 improvement in the sensitivity of the measurements. Multi-pulse integration will provide additional improvements and measurements of ambient concentrations of SO_2 and O_3 appear feasible.

3. Investigation of DIAL in the Infrared Spectral Region

It is important to pursue DIAL in the infrared spectral region because of the large number of species that can be measured in the infrared. Also, in most cases high sensitivity can be achieved. Two limitations appear in the infrared: (1) the smaller backscatter coefficient from the natural atmosphere and less sensitive detectors yield a smaller signal-to-noise ratio (SNR) for the same transmitted energy and receiver aperture, and (2) the potential for spectral interference by water vapor and other species is greater. The first disadvantage is counterbalanced by the larger output from infrared lasers, especially the CO_2 and DF lasers. This larger output improves performance considerably. The second disadvantage can be overcome in almost all cases by a careful selection of the laser wavelengths. In the vast majority of species that we have examined wavelengths could be chosen to yield a small interference, and high-sensitivity measurements could be made on interference-free wavelengths.

The infrared research done at SRI used discretely-tunable lasers, and that at Stanford used continuously-tunable lasers. Discretely-tunable lasers emit laser radiation on many fixed wavelengths. The utility of discretely-tunable lasers for remote measurement depends on the

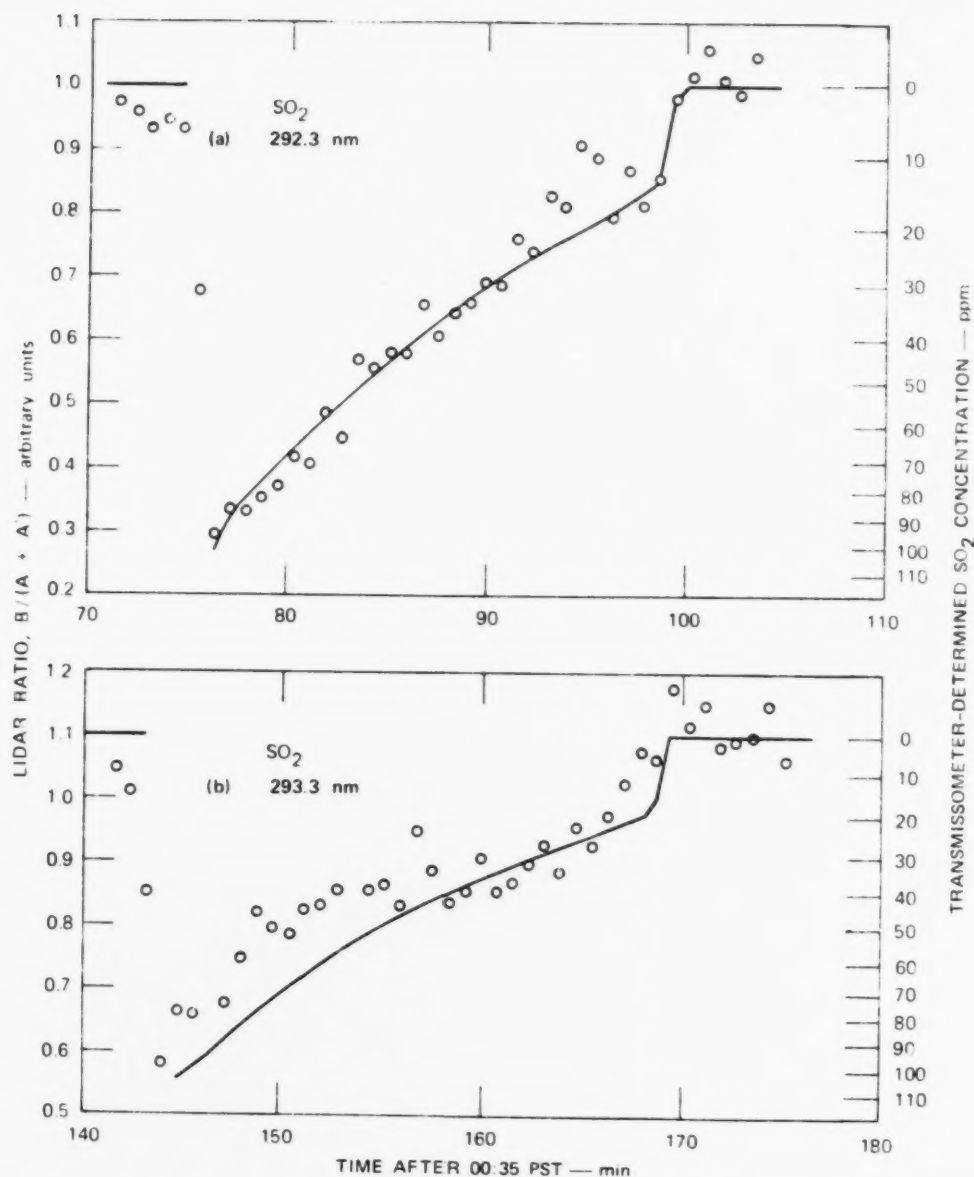


FIGURE 4 PLOTS OF LIDAR RATIO $B/(A+A')$ AS A FUNCTION OF TIME FOR INJECTION OF 110 ppm SO_2 INTO THE SAMPLE CHAMBER. The ratio is normalized to 1 for air in the chamber at 100 min. The ppm scale on the right-hand side of each plot was calculated using Beer's law and the ratio for air in the sample chamber after the SO_2 was exhausted. The solid line is the transmissometer-determined value of the concentration; the points are ratios from an eight-pulse lidar average.

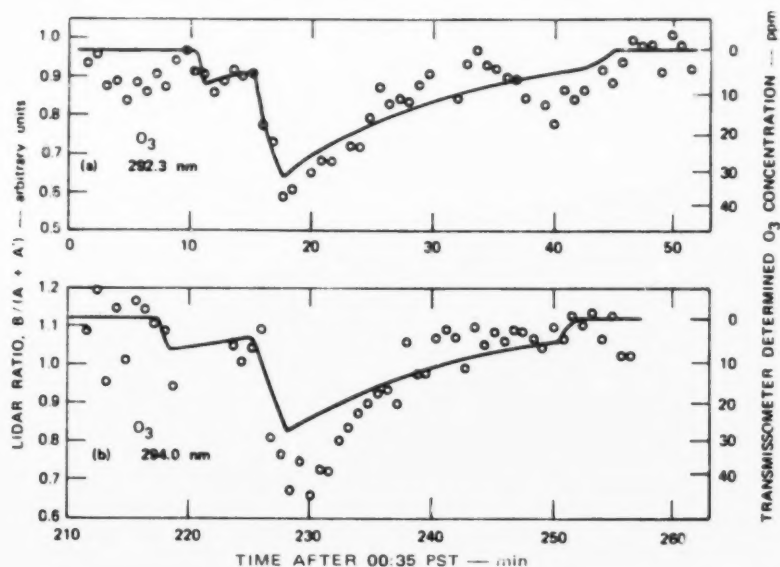


FIGURE 5 PLOTS OF LIDAR RATIO vs TIME WITH O_3 INTRODUCED INTO THE SAMPLE CHAMBER. Time, ratio, and transmissometer concentration determined as for SO_2 in Figure 4.

spectral coincidence of these fixed wavelengths with absorption features of gases of interest. Because of the high spectral density of both fixed wavelength laser lines and absorption lines of pollutants, many useful spectral overlaps exist. Several gas lasers emit high-energy pulses that enable long-range measurements to be made. Hence, gas lasers are useful in making range-resolved measurements using backscatter from naturally occurring aerosols, and integrated concentration measurements using scatter from topographic targets. Continuously-tunable lasers have greater flexibility in the selection of interference-free wavelengths, but the discretely-tuned lasers generally have larger output. Thus, each has advantages and disadvantages, and both were examined in this study because they both appear very promising for remote monitoring.

4. Measurements of HCl, CH₄, and N₂O in the Mid-Infrared

The measurements made by SRI in the mid-infrared involved path-averaged measurements using topographic backscatter. Since we were limited to the available hardware, range-resolved measurements were not attempted. However, performance predictions were made to establish the capabilities of a system comprised of commercially-available components.

A diagram of the single-ended DF laser system used to monitor HCl, CH₄, and N₂O is shown in Figure 6. This system, reported by Murray et al. (1976a) used a topographic target to provide the backscatter signal and measured the integrated concentration over the path between the lidar and the target. The DF laser beam was transmitted coaxially with the receiver and was pointed through a sample chamber that was positioned 300 m away. The backscattered signal from the topographic target was collected by a Newtonian telescope and focused on an infrared detector. The detector signal was amplified and displayed on a chart recorder. Different concentrations of the gases were injected into the sample chamber. The concentrations were measured with both the lidar and an in situ monitor and the values compared.

Experimental parameters are shown in Table 3. The DF laser operated at between 100- and 150-mJ/pulse, although some variations occurred from line to line and day to day. The HgCdTe detector was designed for use near 10 μm , and its detectivity at 3.7 μm was degraded somewhat to $5 \times 10^9 \text{ cmHz}^{1/2} \text{ W}^{-1}$. Detectors were available that are twenty times more sensitive in the 3- to 5- μm region. Nonetheless, the HgCdTe detector was used because it was readily available. Even with the use of a suboptimum detector, system performance was not limited by detector noise. The laser wavelengths and absorption coefficients that were used are shown in Table 4. The wavelengths used for the three gases were between 3.6 and 3.9 μm . The gaseous absorption coefficients at the available DF laser wavelengths result from spectral coincidences. The HCl absorption coefficient was measured by Bair and Allario (1976) and those for CH₄ and N₂O were measured by Spencer et al (1974). The wavelengths were established by Rao (1976).

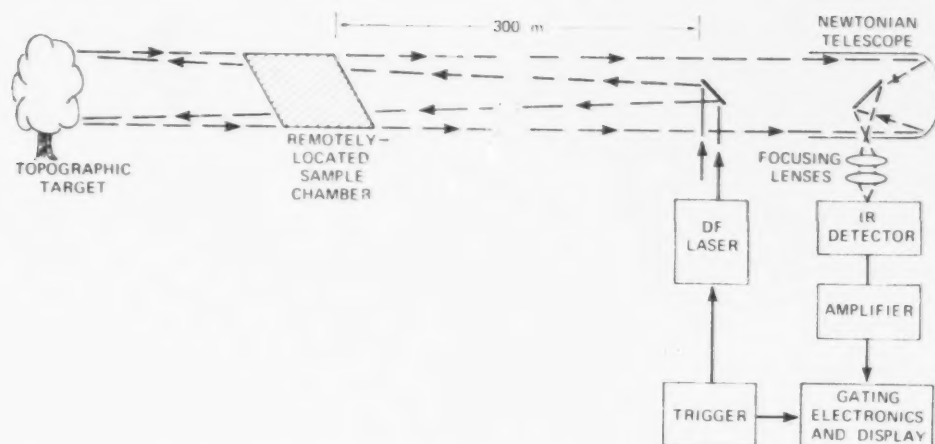


FIGURE 6 EXPERIMENTAL APPARATUS FOR REMOTE MEASUREMENT OF HCl, CH₄, AND N₂O USING A DF LASER

Table 3

EXPERIMENTAL PARAMETERS FOR THE MEASUREMENT OF HCl, CH₄, AND N₂O

<u>DF Laser Transmitter</u>	
Energy	100 to 150 mJ/pulse
Pulsewidth	1.0 μ s (FWHM) [†]
Beam divergence	1.0 mrad (FWHM) [†]
Typical PRF	1/6 Hz
<u>Receiver</u>	
Telescope diameter	31.75 cm
Field of view	3.0 mrad (FWHM) [†]
HgCdTe detector	
D $\cdot$ (3.7 μ m)	$5(10^9) \frac{\text{cm Hz}^{1/2}}{\text{W}}$
Size	1 $\times$ 1 mm
Time constant	75 ns (to 1/e point)

[†] Full width at half maximum.

Table 4

LASER WAVELENGTHS AND ABSORPTION COEFFICIENTS

Gas	DF Laser Line	Wavelength (μ m)	Absorption Coefficient (cm ⁻¹ atm ⁻¹)
HCl	P ₂ (3)	3.636239	5.64
CH ₄	P ₁ (9)	3.715252	0.047
N ₂ O	P ₃ (7)	3.890259	1.19

A typical display of raw data for the HCl measurements is shown in Figure 7. The ratio of the backscattered to the transmitted signal is shown as a function of pulse number. Initially, with no HCl in the chamber, the baseline level of the ratio was established. This was followed by two injections of 173 ppm of HCl . Each injection yielded an easily discernible decrease in the plotted ratio. Near pulse number 127, the sample chamber ports were opened and a fan purged the system of HCl . The continuously increasing ratio during the purge verified the decreasing HCl concentration. Ultimately, the ratio achieved a constant value consistent with the initial level with no HCl in the chamber.

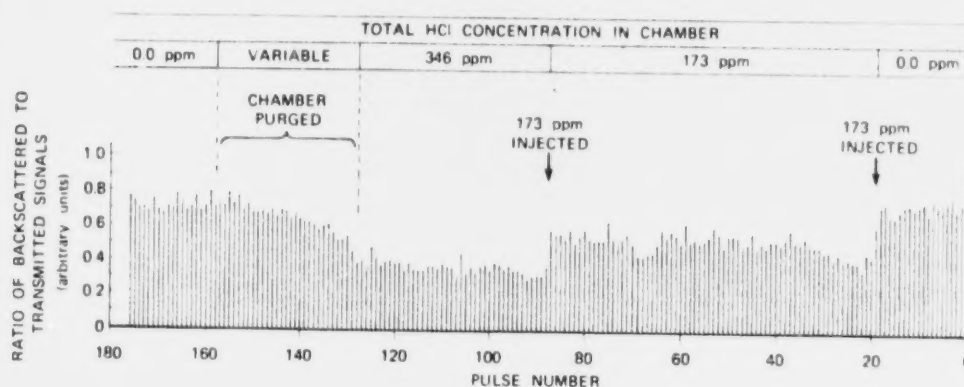


FIGURE 7 SAMPLE OF BEHAVIOR OF THE RATIO OF BACKSCATTERED TO TRANSMITTED SIGNAL DURING SYSTEM OPERATION ON HCl

A summary of the HCl test for different concentrations is shown in Figure 8. The lidar-measured HCl concentration is plotted as a function of the syringe-injected value. The solid line represents perfect agreement between the lidar and the syringe values. Each represents the average of 15 measurements. Each error bar shown represents the standard deviation (SD) for 15 data points. Generally, good agreement is apparent between the lidar and the syringe values, thus confirming that the absorption coefficient used in the data reduction is accurate. The right vertical axis shows the equivalent product of concentration and path length for the individual measurements. The sensitivity of the system, or minimum detectable concentration, is defined by the concentration that

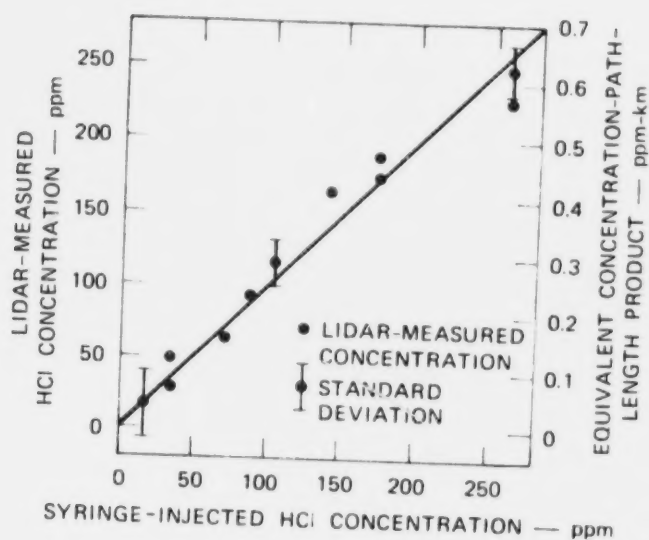


FIGURE 8 SUMMARY OF REMOTE MEASUREMENT TESTS OF HCl IN A SAMPLE CHAMBER

equals the SD. For HCl, the sensitivity was determined to be 0.05 ppm over a 1-km path, or 50 ppb over 1 km.

A summary of the CH_4 data is shown in Figure 9. Again, good agreement was obtained between the lidar and the in situ value, thereby verifying the absorption coefficients used in the data reduction. For CH_4 , the system sensitivity was found to be 6 ppm over a 1-km path.

A summary of the N_2O data is shown in Figure 10. The triangles represent data obtained with a 1-m diameter juniper tree used as the backscattering target; the circles represent data taken using a plywood board. The two targets were used to demonstrate system operation on two different materials with different reflectiveness. The foliage target was used because it provided a reflectivity typical of field operation. Both targets were placed immediately behind the sample chamber. The lidar results obtained with both targets agree with the in situ values. The system sensitivity for N_2O was determined to be 0.24 ppm-km.

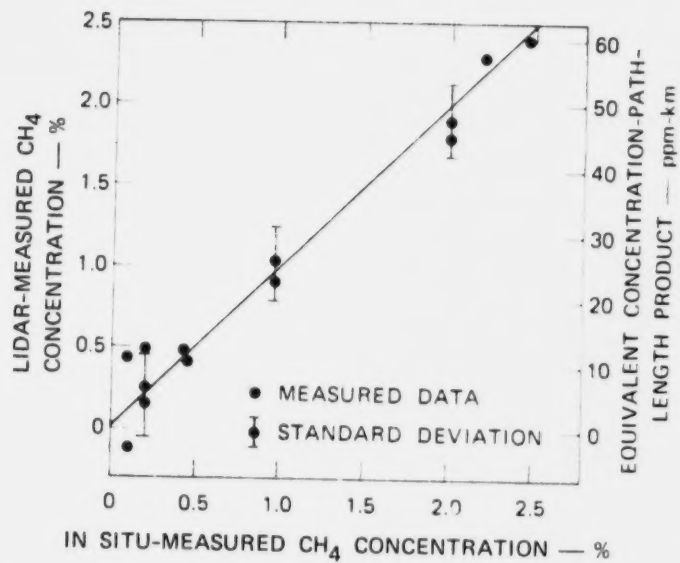


FIGURE 9 SUMMARY OF REMOTE MEASUREMENT TESTS OF CH_4 IN A SAMPLE CHAMBER

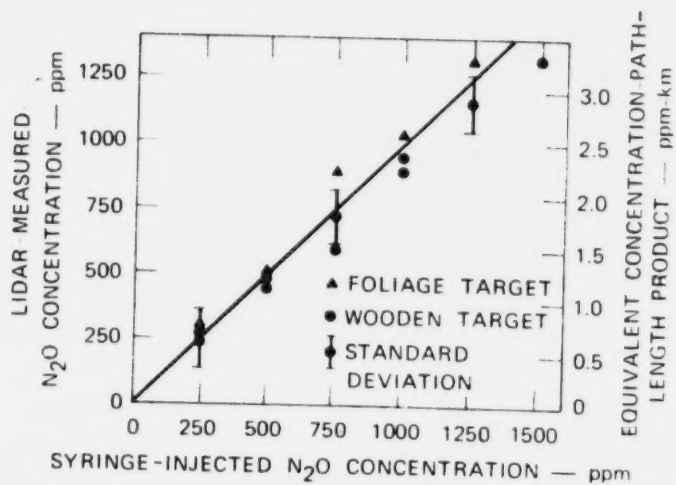


FIGURE 10 SUMMARY OF REMOTE MEASUREMENT TESTS OF N_2O IN A SAMPLE CHAMBER

A plume of N_2O was generated in front of the foliage target to represent typical field conditions. The results of that measurement are shown in Figure 11. The product of N_2O concentration and path length is plotted as a function of the plume generator state, either "off" or "on". The center bar represents the mean value of 15 pulses. The extent of the bar above and below the mean represents 1 SD of those pulses. The increased SD with the generator "on" indicates that turbulent transport was convecting the plume into and out of the lidar line of sight. The increased average value of the product of concentration and path length is a clear indication that the lidar measured the gas concentration in the plume, even though the concentration was only slightly above the estimated system sensitivity of 0.24 ppm-km.

In general, gaseous species in the atmosphere, such as water vapor and HDO , can produce errors in two wavelength differential absorption measurements. The error due to interferences is shown in Table 5 in ppm-km. These data were calculated using an 8-km horizontal path from the data reported by Spencer et al. (1974). The second column represents the measurement error without interference compensation. An arbitrary selection of the weakly absorbed line was used, and this resulted in the measurement error shown in the second column. By carefully selecting the weakly absorbed line so that its transmission by interfering species was matched to that of the strongly absorbed line, the measurement error was reduced to the values shown in the third column. Further interference reduction can be obtained by estimating or measuring the effects of the interfering species at the wavelengths selected. By estimating interference effects to within only 30% of their actual value, the measurement errors shown in the fourth column were calculated. The required sophistication of the interference compensation for a given measurement depends on the application and the desired sensitivity. Table 5 illustrates the use of two techniques for interference minimization. Further reduction in measurement error can be obtained by measuring the concentration of the interfering species. In principle, a negligible measurement error can be obtained using this technique.

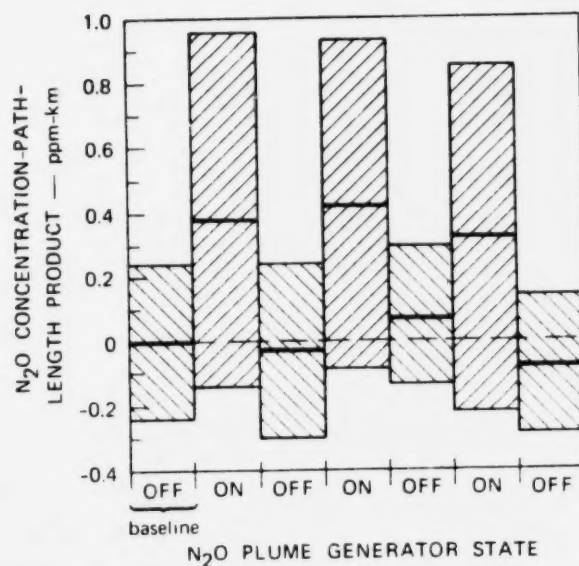


FIGURE 11 RESULTS OF N₂O PLUME MEASUREMENT IN THE OPEN ATMOSPHERE NEAR THE LIMIT OF DETECTABILITY OF THE SYSTEM

Table 5

MEASUREMENT ERROR (ppm-km) DUE TO INTERFERING SPECIES OVER AN 8-km HORIZONTAL PATH^(a)

Target Gas	No Interference Compensation ^(b)	Line Selection to Minimize Interferences ^(c)	Interference Compensation ^(c,d)
HCl	0.10	0.012	0.0037
CH ₄	10.0	2.0	0.59
N ₂ O	0.73	0.041	0.012

- (a) Errors calculated from data of Spencer et al. (1974).
 (b) DF laser lines (strongly absorbed/weakly absorbed) for HCl, CH₄, and N₂O are, respectively, P₂(3)/P₁(6), P₁(9)/P₂(5), and P₃(7)/P₁(14).
 (c) DF laser lines (strongly absorbed/weakly absorbed) for HCl, CH₄, and N₂O are, respectively, P₂(3)/P₃(6), P₁(9)/P₁(10), and P₃(7)/P₂(10).
 (d) Compensation is calculated from an estimate of absorption by interfering species. The ratio between the transmission of the strongly absorbed laser lines and that of the weakly absorbed laser lines is assumed to be accurate to within 30% of the actual value.

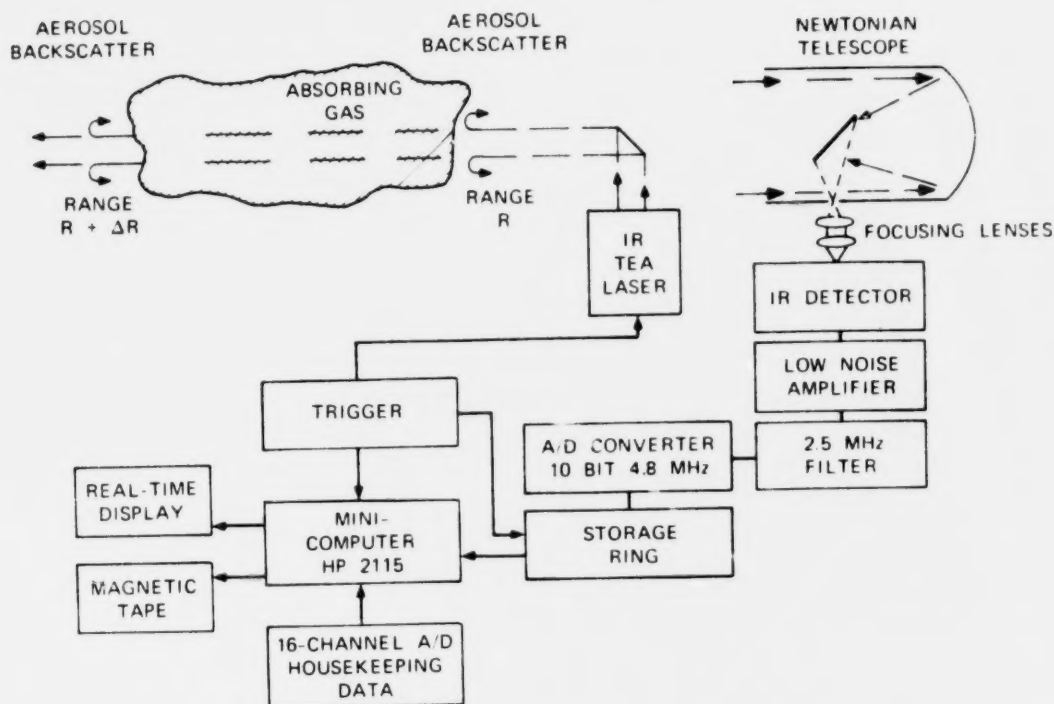


FIGURE 12 THE INFRARED DIFFERENTIAL ABSORPTION LIDAR SYSTEM FOR RANGE-RESOLVED MEASUREMENT OF GASEOUS SPECIES

coaxially with the receiving telescope. Scattered radiation from naturally-occurring aerosols was collected by the telescope and focused on a HgCdTe detector. The detector signal was amplified, filtered, digitized, displayed, and stored on magnetic tape for later analysis. A real-time display of the return signal on the absorbed and nonabsorbed laser lines was used to monitor system operation.

The system parameters are shown in Table 6. The laser operation on these strong lines was quite stable with 90% of the pulses falling within $\pm 5\%$ of the average energy. The field of view of the receiver was designed to be nearly twice the beam divergence of the laser to ensure easy alignment of the system and to minimize the influence of thermal drift during an experiment. Three detector configurations were used during the investigation. At first, a very high sensitivity detector was used because of uncertainty about the magnitude of the backscatter coefficient. This

Table 6
DIAL SYSTEM PARAMETERS

<u>CO₂ TEA Laser</u>							
Transmitted energy	1 J						
Lines	R(12), R(18), R(20) on 10-μm band						
Pulse width	100 ns (FWHM)						
Beam divergence	1.8 mrad (to the 10% point)						
Typical PRF	0.5 Hz						
<u>Receiver</u>							
Telescope diameter	31.75 cm						
Field of view	3.3 mrad (to the 10% point)						
Hg-Cd-Te detectors							
	<table><tr><td><u>Detector 1</u></td><td><u>Detector 2</u></td><td><u>Detector 3</u></td></tr><tr><td>$2(10^{11}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$</td><td>$2(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$</td><td>$1.1(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$</td></tr></table>	<u>Detector 1</u>	<u>Detector 2</u>	<u>Detector 3</u>	$2(10^{11}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$	$2(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$	$1.1(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$
<u>Detector 1</u>	<u>Detector 2</u>	<u>Detector 3</u>					
$2(10^{11}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$	$2(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$	$1.1(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$					
D*							
Time constant	3.0 μs						
Size	1 x 1 mm						

detector has a time constant of 3 μ s, which degraded range resolution. After a large backscattered signal was obtained, the cooled filter was removed, decreasing the time constant to 0.98 μ s and increasing the detector noise by a factor of 10. To improve range resolution further, a faster detector was obtained having a time constant of 0.075 μ s.

The first target species for the CO₂ lidar system was water vapor, and considerable system optimization was performed on this species. The water vapor absorption coefficients measured by Shumate et al. (1976) are shown in Table 7. These indicate that the R(20) transition is more strongly absorbed than either the R(12) or the R(18). Most of the results were obtained using the R(20) and R(18) laser lines. The ambient conditions on the days of the measurement included a visibility of 16 to 24 km and water vapor content in the neighborhood of 0.5 to 1.0%.

Figure 13 shows typical lidar return signals that were obtained with the 0.98- μ s detector. The sharply rising aerosol return results from the geometric overlap of the transmitter and receiver. Following system convergence, the $1/R^2$ and atmospheric attenuating losses decrease the signals, which drop into the noise between 1.5 and 2.0 km. The

Table 7

WATER VAPOR ABSORPTION COEFFICIENTS

Line	λ (μm)	α ($10^{-4} \text{ cm}^{-1} \text{ atm}^{-1}$)
R(12)	10.303 458	1.60
R(18)	10.260 381	0.935
R(20)	10.246 625	8.65

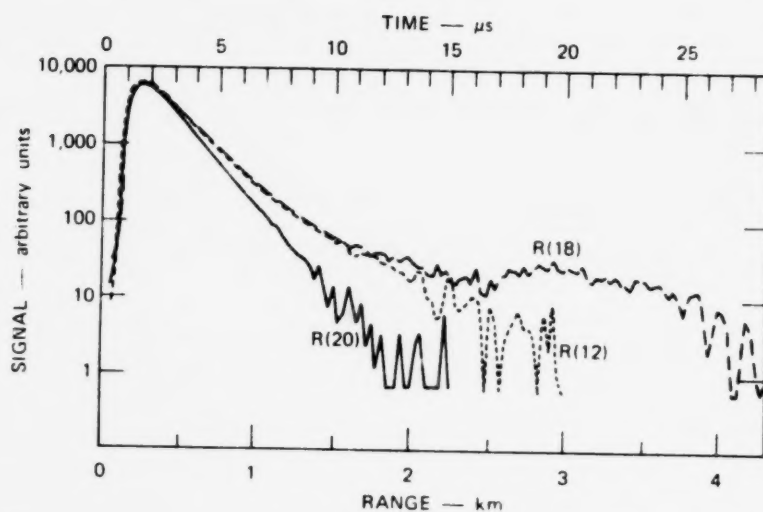


FIGURE 13 SIGNAL BACKSCATTERED FROM ATMOSPHERIC AEROSOLS FOR THE R(12), R(18), AND R(20) LINES ON THE 10- μm BAND OF CO_2 . The R(20) line is more strongly absorbed by ambient H_2O than is the R(12) or R(18). The 0.98- μs detector was used for these results.

differential between the R(20) and the R(18) and the R(12) lines is readily apparent. The rate of change of the differential absorption between these lines is proportional to the concentration of the absorbing species.

The water vapor concentration was calculated using the data of Figure 13. The resulting water vapor profile is shown in Figure 14 as a function of range. The point monitor was a recording hygrothermograph located on the roof of a building at a range of 300 m near the lidar line of sight. Good agreement was obtained between the point monitor and the lidar values for short ranges. The lidar-determined concentration appeared to undergo random fluctuations at less than 1 km, whereas the return signal was relatively free of noise to a range of 1.5 km. This is indicative of the propagation of noise effects from the return signal through the DIAL equation. This demonstrates the distinction between the differential SNR and the SNR of the return signal.

Since the above results were obtained, two major changes have significantly improved system performance. The installation of a fast detector with a 75-ns time constant improved the range resolution, and better receiver alignment increased the system range. The results shown in Figure 15 were obtained with these modifications. The signal did not drop into the noise until a range of a factor of 2 over the results obtained earlier. The error bars shown in Figure 15 represent the uncertainties for a single pulse and also those for the 200 pulses that were summed to obtain the curve. The single pulse uncertainty is the SD of the return signals at each range. The multipulse uncertainty is the single pulse uncertainty divided by the square root of the number of pulses. The increasing uncertainty as a function of range indicates the decreasing SNR.

The concentration derived from the new data is shown in Figure 16. The uncertainties associated with the return signal are propagated through the data reduction process. The resulting uncertainties in concentration were calculated using the standard theory presented by Beers (1962) and are shown in Figure 16. In this case, the increasing uncertainty in the multipulse measurement equaled the measured concentration near 1.5 km. This range represented the maximum range for which the measured values have significance. The negative concentration values have no significance in reality and are displayed to illustrate how the measurement degrades with increasing range.

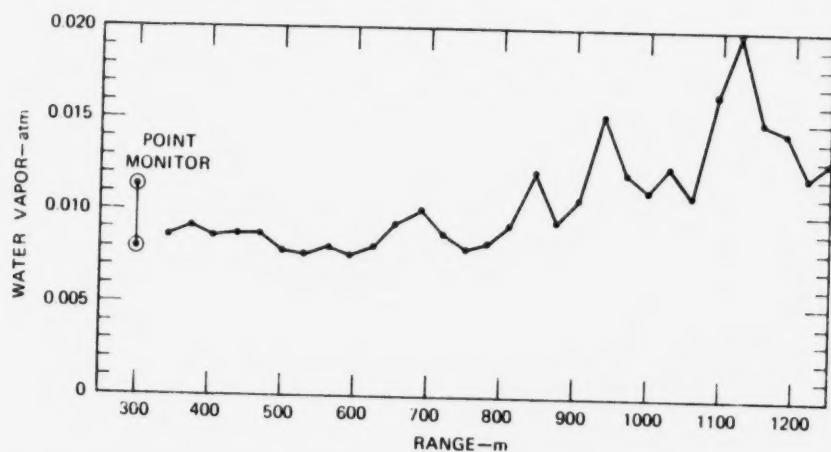


FIGURE 14 WATER VAPOR CONCENTRATION MEASURED USING THE CO₂ LIDAR. A 60-m range cell was used in the data reduction. The point monitor was a calibrated recording hygrothermograph located near the lidar line of sight.

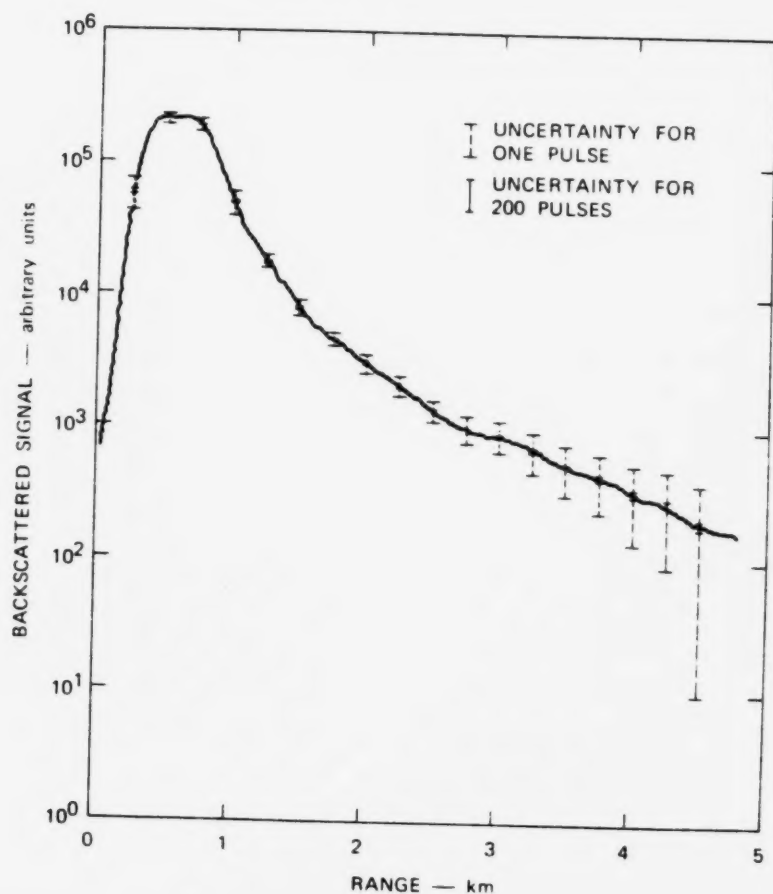


FIGURE 15 TYPICAL RETURN SIGNAL SUMMED OVER 200 PULSES SHOWING UNCERTAINTIES AS A FUNCTION OF RANGE

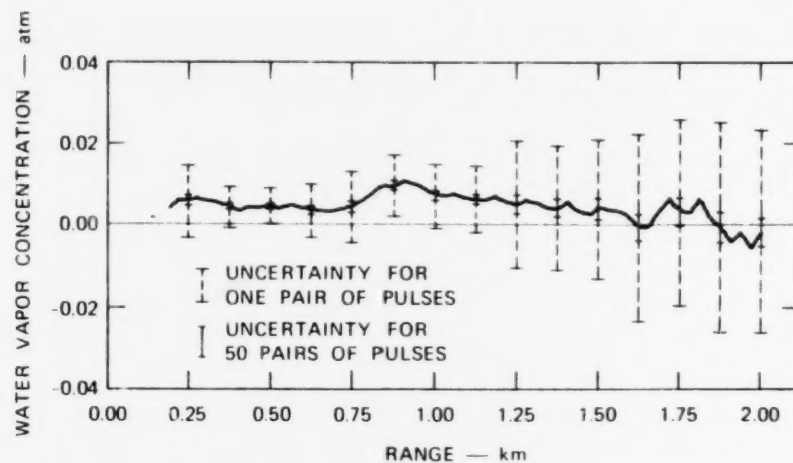


FIGURE 16 RANGE-RESOLVED WATER VAPOR CONCENTRATION AND UNCERTAINTIES AS A FUNCTION OF RANGE

In a prediction of the range-resolved performance of the CO_2 lidar system, the following parameters were used:

$$E = 15 \text{ J pulse}^{-1}$$

$$A_r = 1.17 \text{ m}^2$$

$$\lambda = 10.0 \text{ } \mu\text{m}$$

$$K = 0.5$$

$$\beta_{180} = 1.2 \times 10^{-7} \text{ m}^{-1} \text{ sr}^{-1}$$

$$\alpha = 0.0927 \text{ km}^{-1}$$

$$D^* = 2 \times 10^{10} \text{ cmHz}^{1/2} \text{ W}^{-1}$$

The backscatter coefficient is characteristic of a low-altitude haze and is somewhat less than it is in the 3- to 4- μm region. The detector characteristics are those consistent with available detectors. The calculation of SNR yields a value of 50 at a range of 10 km for a single pulse. Thus, high sensitivity, range-resolved measurements are feasible at 10 km. Longer range or higher sensitivity can be obtained using multi-pulse integration.

The sensitivity of the CO₂ lidar system to different gases has been calculated based on an SNR of 200. The sensitivity values for 16 representative compounds are shown in Table 8. The absorption coefficients vary widely. Those for CO₂ and water vapor are relatively small. This is necessary to permit long-range transmission of the beam. The absorption coefficient of many other gases is high, yielding a sensitivity of 0.063 to 22.0 ppb-km. Range-resolved multigas monitoring of these gases appears feasible at a 10-km range with commercially available components.

Table 8

CALCULATED CO₂ LIDAR SYSTEM SENSITIVITIES

Species		Absorption Coefficient (cm ⁻¹ atm ⁻¹)	Sensitivity (ppb-km)
Name	Formula		
Ammonia	NH ₃	120.0	0.42
Benzene	C ₆ H ₆	2.3	22.
1,3 butadiene		3.45	15.
Carbon dioxide	CO ₂	1.8 (10 ⁻³)	28,000.
Ethylene	C ₂ H ₄	33.0	1.5
Freon 113	C ₂ Cl ₃ F ₃	19.2	2.6
Freon 11	CCl ₃ F	31.0	1.6
Freon 12	CCl ₂ F ₂	92.0	0.55
Methanol	CH ₃ OH	19.4	2.6
Ozone	O ₃	12.7	4.0
Perchloroethylene	C ₂ Cl ₄	28.5	1.8
Sulfur dioxide	SO ₂	6.73	7.5
Sulfur hexafluoride	SF ₆	800.0	0.063
Trichloroethylene	C ₂ HCl ₃	14.0	3.6
Vinyl chloride	C ₂ H ₃ Cl	6.79	7.4
Water vapor	H ₂ O	8.36 (10 ⁻⁴)	60,000.

From Table 8 it is evident that line-tunable infrared laser sources have adequate spectral coincidences to allow measurements of a number of important molecular species. The other advantages of these sources of high pulse energy, commercial availability, and relatively low cost, make them promising for future remote monitoring applications.

6. Path-Averaged Measurements of Gases Using a DIAL System With CO₂ Laser Transmitter

Path-averaged measurements can be performed simultaneously with range-resolved measurements by pointing the lidar at a topographic target. Figure 17 shows an example of a return signal on the P(14) line of the 10- μ m band of CO₂ with the lidar pointed west toward the coastal foothills near Menlo Park, California. A well-defined topographical reflection primarily from foliage at 10 km was obtained. In such a configuration, the range-resolved concentration of a pollutant can be obtained to a range limited by the SNR. Simultaneously, the path-averaged concentration of the pollutant over a longer range between the lidar system and an arbitrary topographical target can be obtained. Wide applicability of such combination range-resolved and path-averaged lidar systems is foreseen for use in source and ambient air quality monitoring.

A diagram of the CO₂ lidar system used to make path-averaged measurements of ethylene is shown in Figure 18. The lidar was pointed toward the coastal foothills, which are 5 km south of the SRI site. A topographic target, consisting primarily of foliage, scattered radiation that was collected by the Newtonian telescope. Focusing lenses were used to expand the field-of-view of the receiver so that it was larger than the beam divergence of the laser. The detector signal was amplified and then filtered to broaden the return pulse. A beamsplitter was used to divert a portion of the transmitted beam to a pyroelectric detector that monitored the energy of the transmitted pulse. Both the backscattered and transmitted pulses were integrated to yield dc signals. The gate time of the integrators was set at 1 μ s. The time delay of the gate was set so that the gate was centered on the pulse. The ratio of the backscattered to transmitted signal was obtained using an analog divider. This ratio was displayed on a chart recorder.

The system parameters are shown in Table 9. The laser had a pulse-to-pulse stability such that 90% of the pulses were within $\pm 5\%$ of the average energy. The P(14) and P(16) lines have absorption coefficients of 29.10 and 4.55 cm⁻¹ atm⁻¹, respectively, as reported by Patty et al. (1974).

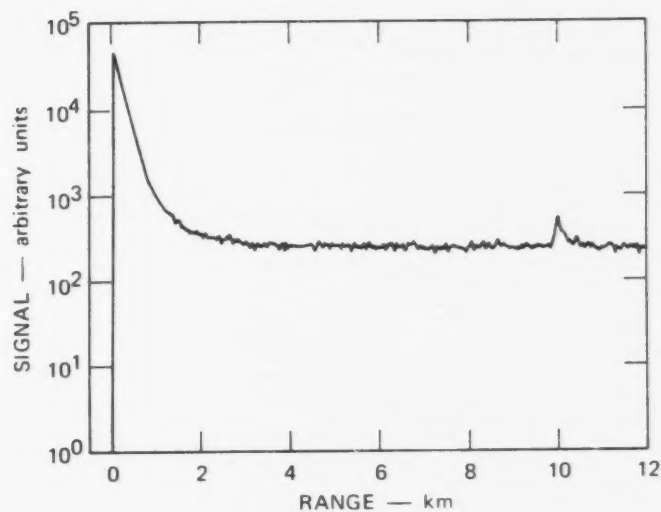


FIGURE 17 BACKSCATTERED SIGNAL INCLUDING ATMOSPHERIC RETURN AND SCATTER FROM THE COASTAL FOOTHILLS AT A RANGE OF 10 km

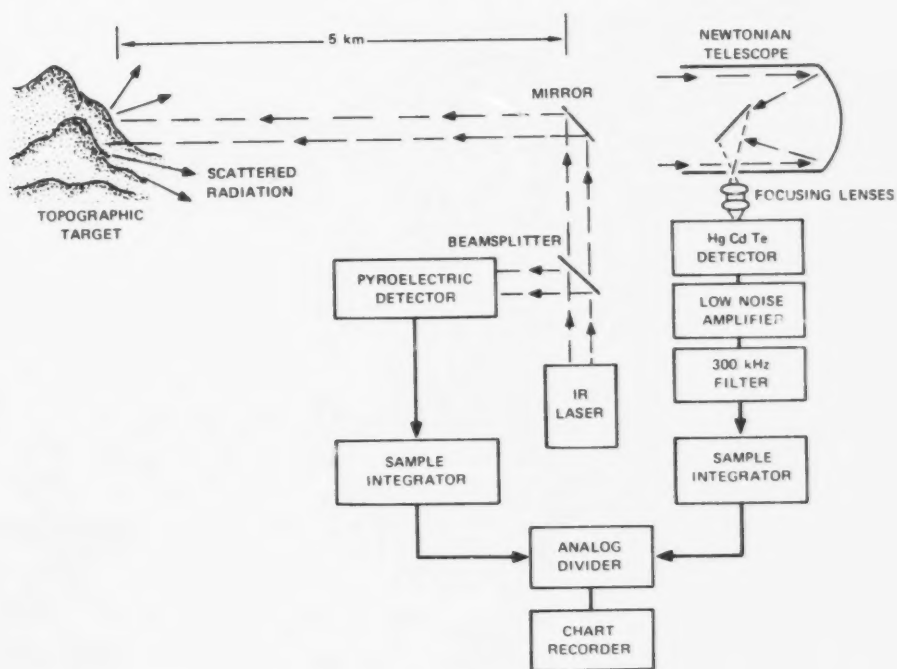


FIGURE 18 INFRARED LIDAR SYSTEM USED TO MAKE PATH-AVERAGED MEASUREMENTS OF ETHYLENE

Table 9

SYSTEM PARAMETERS FOR THE MEASUREMENT OF ETHYLENE

CO₂ TEA Laser

Transmitted energy	1 J
Lines	P(14) and P(16)
Frequency	949.479 and 947.742 cm ⁻¹
Pulsewidth	100 ns (full-width half maximum)
Beam divergence	1.8 mrad (to the 10% point)
Typical PRF	0.5 Hz

Receiver

Telescope diameter	31.75 cm
Field of view	3.3 mrad (to the 10% point)
Hg-Cd-Te detector	
D*	$1.1(10^{10}) \frac{\text{cm Hz}^{\frac{1}{2}}}{W}$
Time constant	0.075 μ s
Size	1 X 1 mm

An example of the backscattered signal obtained with the infrared lidar system is shown in Figure 19. The signal backscattered from aerosols decreases with increasing range. Near 5 km in range, it falls into the system noise. The backscatter from the foothills at a range of 5 to 6 km is approximately 300 times larger than the sum of the noise and the aerosol backscatter at that range.

A sample of data representing the ratio of the topographically scattered signal to the transmitted signal is shown in Figure 20 for the P(14) and P(16) laser lines. The P(14) line is more strongly attenuated by ambient ethylene because its absorption coefficient is larger. The SD of data within each group averages 3%. The system noise is due to occasional laser arcing, movement of the foliage target in the wind, detector noise, atmospheric scintillation, and variations in aerosol scattering. The average standard deviation of 3% is equivalent to a

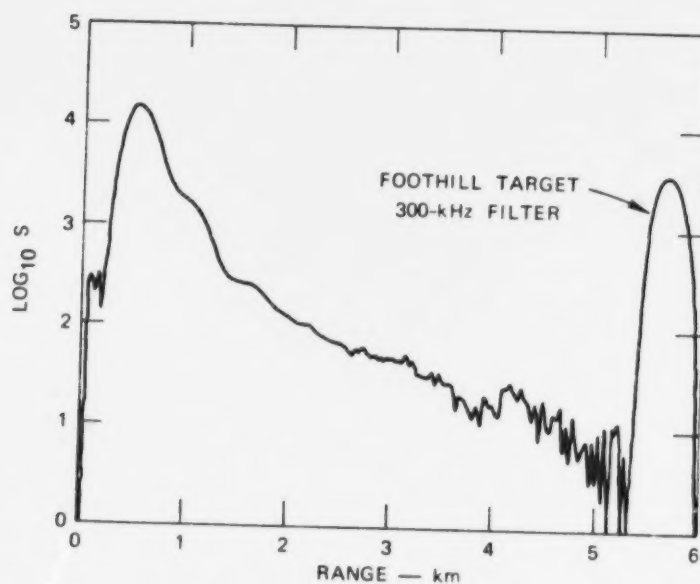


FIGURE 19 BACKSCATTERED SIGNAL MEASURED WITH THE INFRARED LIDAR SYSTEM

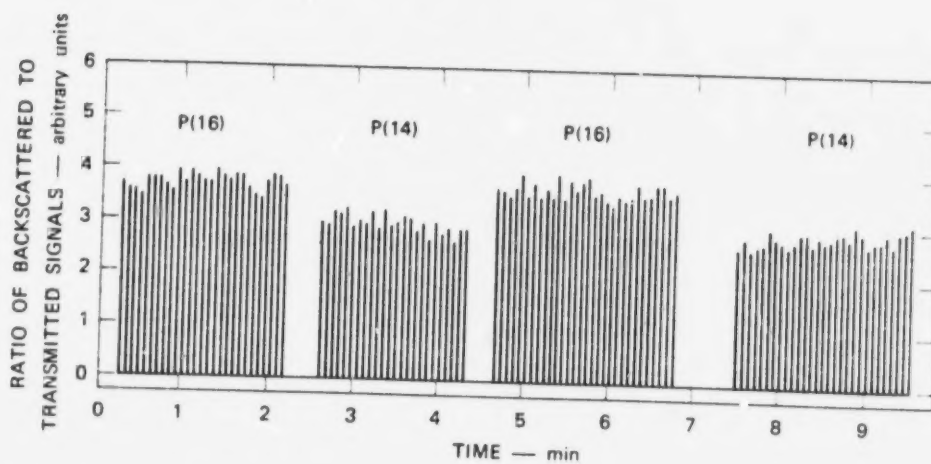


FIGURE 20 RATIOS OF BACKSCATTERED TO TRANSMITTED SIGNAL OBTAINED ON DAY OF HIGH ETHYLENE CONCENTRATION

measurement uncertainty of 0.4 ppb of ethylene when averaging 25 pulse-pairs. The data of Figure 20 were taken on a day during which the concentration of ambient ethylene averaged 20 ppb.

Data taken during a period for which the ethylene concentration averaged 2 ppb are shown in Figure 21. Note that the P(16) line is more strongly absorbed over the 5-km path than the P(14) line. This is the opposite of what would be expected from the relative magnitude of the absorption coefficients due to ethylene. In the higher concentration case shown in Figure 20, this anomalous absorption is masked by the large absorption due to ethylene. We conclude that an interfering species is causing this offset.

To investigate this interference problem a synthetic transmission spectrum was generated as shown in Figure 22. The calculations use a data base similar to that of McClatchey et al. (1973). Calculations were made over a 10-km horizontal path assuming a water vapor concentration consistent with that found on the day of the measurements. The composition of the synthetic atmosphere used for the computer calculations includes the following quantities: $\text{H}_2\text{O} = 0.00657 \text{ atm}$, $\text{CO}_2 = 325 \text{ ppm}$, $\text{O}_3 = 60 \text{ ppb}$, $\text{N}_2\text{O} = 200 \text{ ppb}$, and $\text{NH}_3 = 4 \text{ ppb}$. Ethylene was not included in the calculations. The species producing the largest contribution to the absorption over the frequency interval 994 to 952 cm^{-1} are H_2O and CO_2 .

From Figure 22 the P(14) line has a higher transmission than the P(16) line. Attenuation at these wavelengths is caused by CO_2 and H_2O absorption. Small differences in absorption occur between adjacent CO_2 lines. The P(16) line is located on the edge of a strong H_2O line. Thus, H_2O is the interfering species responsible for the differential absorption of Figure 20. The computer calculations predict the ratio of the transmission on the P(14) line to that on the P(16) line along a 10-km path to be 1.15 for an ethylene-free atmosphere. The ratio of the transmission on the P(14) line to that on the P(16) line through the atmosphere (R) is comprised of a ratio due to ethylene (R) and a ratio due to all other components (R_a) such that $R = R_e R_a$. The value of R_a predicted by the computer along a 10-km path is predicted to be 1.15.

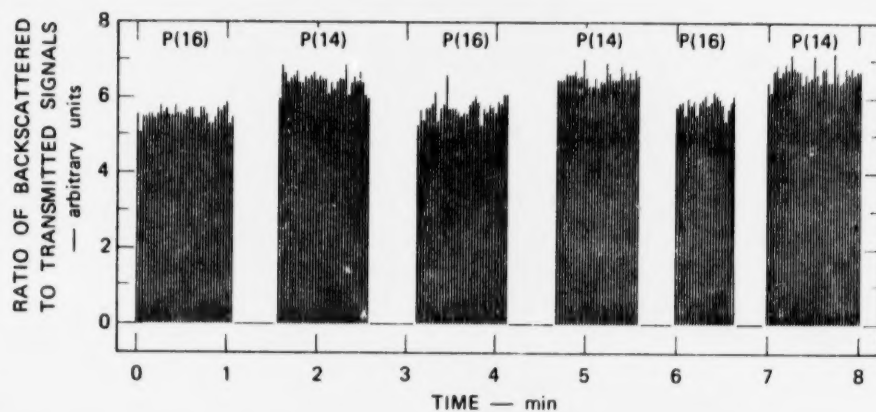


FIGURE 21 RATIOS OF BACKSCATTERED TO TRANSMITTED SIGNAL OBTAINED ON DAY OF LOW ETHYLENE CONCENTRATION

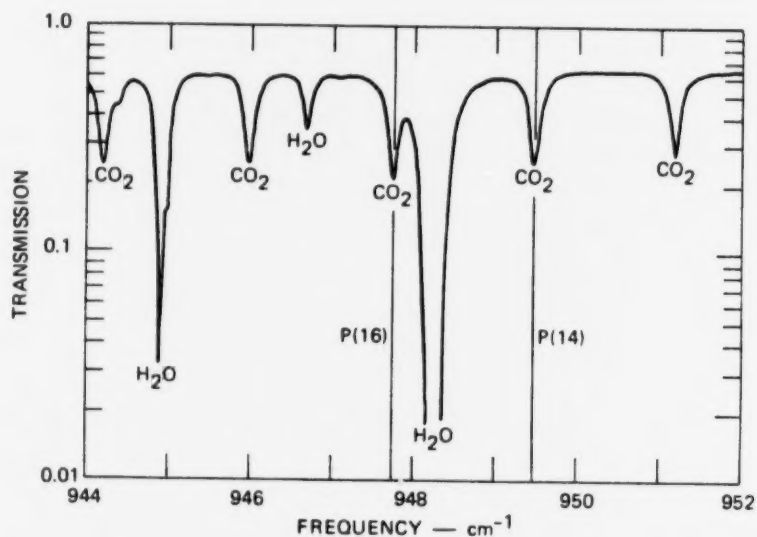


FIGURE 22 SYNTHETIC TRANSMISSION SPECTRUM OF ATMOSPHERE CALCULATED FROM A MODIFIED McCLATCHY DATA BASE

The lidar system measures the value of R . Experimental data on R were obtained on a day in which the ethylene concentration averaged 2 ppb as measured using gas chromatography. The resulting value of R was 1.14. Absorption over the path due to ethylene was calculated assuming a 2-ppb concentration, and we find $R_e = 0.95$. Therefore $R_a = 1.20$, which is in agreement with the calculated value of 1.15. The ratio of 1.20 is equivalent to a systematic error of 7.6 ppb of ethylene. Correction for this systematic error can be made by adding 7.6 ppb to each ethylene measurement. The correction varies with water vapor concentration. For this locale, water vapor typically varies $\pm 20\%$ about its mean during the day. Thus, if the correction factor of 7.6 ppb were applied, we would expect to introduce a random uncertainty of ± 1.5 ppb. If higher sensitivity is required, the water vapor content can be monitored using two different CO_2 laser lines. Then, a more accurate correction factor can be applied to the data. Such a correction process would yield an uncertainty due to water vapor interference of less than 1 ppb.

The results of ethylene measurements with the 7.6-ppb correction included are shown in Figure 23. The two days represent the largest and smallest concentrations that we have measured. Major urban areas can have ethylene concentrations 20 times our maximum [see Abeles and Heggstad (1973)]. The lidar uncertainties represent a combination of the random system uncertainty and the variable component of the water vapor correction described above. The point monitor results were obtained by the gas chromatographic analyses of sealed-syringe samples taken at five ground-level locations near the lidar line of sight. A point-by-point comparison of the lidar and the point monitor data is inappropriate because they measure different quantities. The point monitor data represent the concentration present at a single point in space and time. The lidar measured the concentration integrated over a 5-km path. However, both the lidar and the point monitor should record generally high levels on smoggy days and low levels on clean days. To that extent the data obtained with the two systems are in agreement.

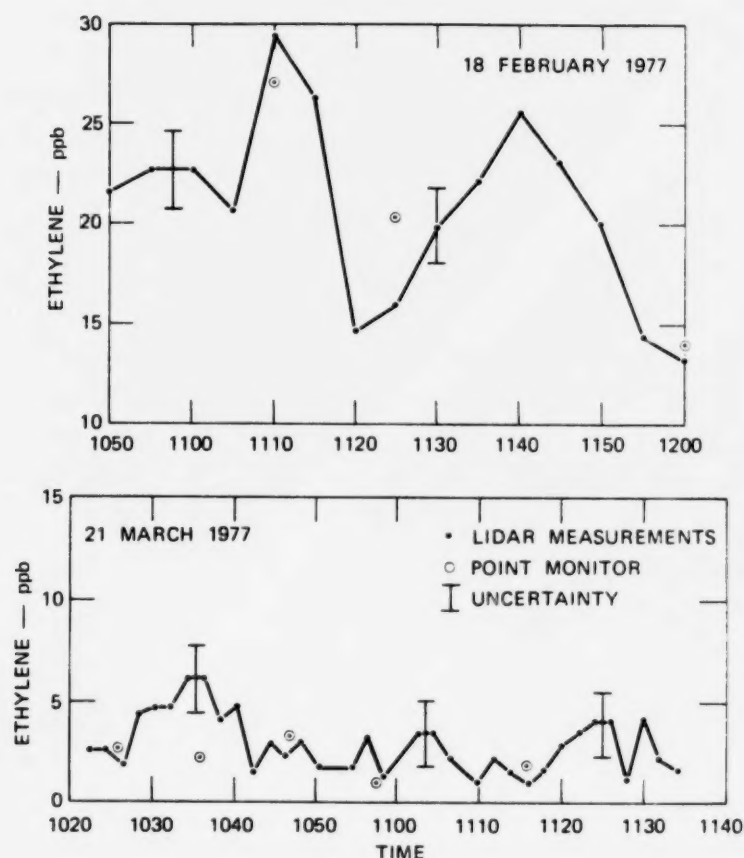


FIGURE 23 MEASURED ETHYLENE CONCENTRATIONS CORRECTED FOR WATER VAPOR INTERFERENCE

7. Measurement of Average Temperature Using a CO₂ Lidar System

The development of a remote measurement capability for temperature has been pursued by many investigators using active and passive techniques and is quite important for meteorological studies and as an input to pollution models and studies of atmospheric chemistry. The passive techniques involve measurement of thermal emissions that are temperature-dependent, such as in the work of Anderson and Griffiths (1977). Active techniques have applied either Raman scattering or the differential-absorption lidar technique. Range-resolved measurements of atmospheric temperature using Raman scattering have been reported by Strauch, Derr, and Cupp (1971); Cooney and Pina (1976); and Gill et al. (1979).

A DIAL system using absorption by the O_2 A-band was proposed and analyzed by Mason (1975) as a technique for profiling atmospheric temperature. Measurements with the DIAL technique on the O_2 A-band were performed by Barton and LeMarshall (1979), who found some correlation between O_2 absorption and atmospheric pressure. They concluded that greater transmitted energies and improved frequency stability would be required to implement an operational system. Kalshoven and Korb (1978) have described a DIAL system that uses the O_2 A-band absorption to measure atmospheric pressure and temperature; the system is expected to be operational soon. Penner (1977) outlined a double-ended laser absorption technique that employs two widely-spaced vibrational-rotational absorption lines in CO to measure mean atmospheric temperature between the earth and a spaceborne platform.

The present work includes both the theory and the measurements obtained in applying a single-ended 10- μ m DIAL system that uses absorption by naturally-occurring CO_2 to make path-averaged measurements of atmospheric temperature. It is important to select fairly well-mixed species such as CO_2 for the temperature-dependent absorber. Two widely-spaced vibrational-rotational lines, the P(38) and P(20) transitions of the 10- μ m band, were chosen for their relative high sensitivity and relative freedom from spectral interference. The transmission of one wavelength relative to the other is shown to be an effective measure of the average temperature over a 5-km path. Using CO_2 lidar and scattering from the foothills located 5 km south of the SRI site, relative transmission over that path was measured. Good correlation was obtained between the lidar-measured temperatures and thermometer-measured values.

The DIAL technique can provide remote measurement of temperature by measuring temperature-dependent transmission in atmospheric species. Two wavelengths are employed to measure the relative transmission in a technique similar to that for the remote sensing of gases, as reported by Murray and van der Laan (1978). The governing relationship is given by:

$$T_{rel} = e^{-2\Delta\alpha CL}$$

where T_{rel} is the transmission on one wavelength divided by that on the other wavelength, $\Delta\alpha$ is the difference between the absorption coefficient at one wavelength and that at the other, C is the concentration of the absorbing species, and L is the path length for a path-averaged measurement or range-resolution for a depth-resolved measurement. To determine the concentration of a gas, one measures T_{rel} and, knowing $\Delta\alpha$ and L , solves for C . To determine temperature, one measures T_{rel} , assumes C is constant at some known value, and solves for $\Delta\alpha$ which is a known function of temperature. To avoid error, the concentration of the absorbing species must be known and spatially uniform; to this end CO_2 was selected because it has a concentration of 330 ppm and is fairly well mixed.

Small systematic variations of atmospheric CO_2 have been well documented by Keeling et al. (1976) and by Bolin and Keeling (1963). The latter presented typical variations between the CO_2 concentrations at the South Pole and at Mauna Loa, Hawaii, of 2 ppm to 3 ppm; they also found typical seasonal variations of 6 ppm at Hilo and Mauna Loa. Keeling et al. (1976) found a 1-ppm-per-year increase in CO_2 content and a diurnal variation of approximately 1 ppm on Mauna Loa. The maximum local contamination due to volcanic emissions was approximately 6 ppm. Most locations that are far removed from such emission sources can probably expect diurnal variations of 1 ppm to 3 ppm, which, if not accounted for, would yield an error in the temperature measurement of $0.12^\circ C$ to $0.36^\circ C$; such variations in CO_2 content will probably not be the limiting error mechanism. If larger variations in CO_2 content occur, a local point monitor should be used to correct the error due to these temporal variations.

The wavelengths for the measurement were carefully selected to provide good sensitivity to temperature, freedom from spectral interference, and good laser performance. Of the many wavelength combinations investigated, the best one consists of the P(38) and P(20) lines of the 10- μm band. Using a modified McClatchey et al. (1973) data base, the difference in the absorption coefficient between these two lines was calculated (Figure 24). Near $20^\circ C$, the slope of the curve is 2.1% per $1^\circ C$; thus, measurement of T_{rel} to an accuracy of 1% would yield an uncertainty in temperature of $0.74^\circ C$.

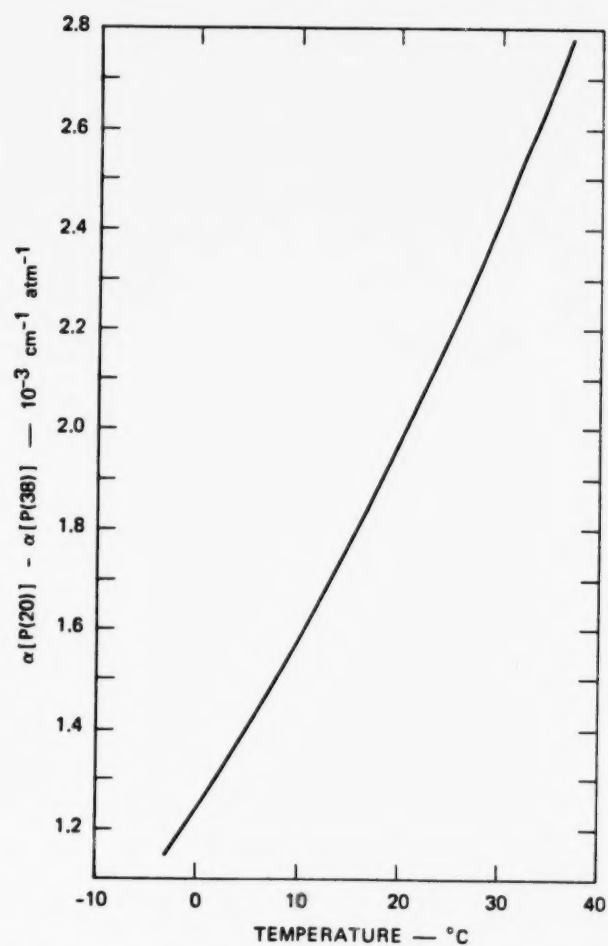


FIGURE 24 CALCULATED DIFFERENTIAL ABSORPTION COEFFICIENT OF CO_2 FOR THE P(20) AND P(38) LINES OF THE $10\text{-}\mu\text{m}$ BAND AS A FUNCTION OF TEMPERATURE

There is some spectral interference caused by atmospheric H_2O . The relative transmission of the P(38) to the P(20) lines for a synthetic atmosphere containing 330 ppm of CO_2 , 50 ppb of O_3 , 200 ppb of N_2O , 4 ppb of NH_3 , 10 ppb of C_2H_4 , and variable amounts of H_2O was calculated (Figure 25). In determining temperature, the error caused by inaccurate knowledge of the H_2O content decreases with increasing temperature. Near $20^\circ C$, the discrepancy between temperatures inferred at $H_2O = 0.006$ atm and at $H_2O = 0.014$ atm is approximately $2.0^\circ C$. If the H_2O content can be estimated within 20% over a horizontal path by a local point monitor, then the error in temperature will be less than $0.5^\circ C$. Since the slope of these curves in Figure 25 is approximately $1.7\%/^\circ C$ near $20^\circ C$, measurement to T_{rel} to an accuracy of 1% would yield an uncertainty in temperature of $0.6^\circ C$. This value represents the sensitivity of lidar-measured temperature to error or small changes in T_{rel} ; it is slightly different from the value calculated from Figure 24 because of the influence of interferences.

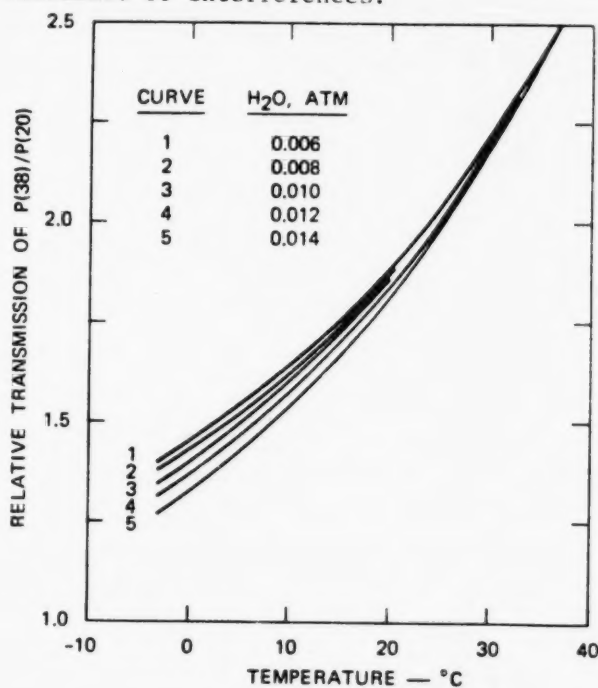


FIGURE 25 CALCULATED RATIO OF TRANSMISSION ON P(38) LINE TO THE P(20) LINE AS A FUNCTION OF TEMPERATURE AND ABSOLUTE WATER VAPOR CONTENT OVER A 10-km HORIZONTAL PATH

The experimental apparatus used to perform these path-averaged measurements of temperature is similar to that shown in Figure 18 except that no filter was used and the sample integrators were replaced by peak-reading voltmeters. The system parameters are given in Table 10. The beam from a tunable, pulsed CO_2 laser was directed coaxially to the receiving telescope. Backscattered radiation from the foothills 5 km south of the SRI site was collected with a Newtonian telescope and focused on an infrared detector. A portion of the transmitted beam was diverted by a beamsplitter to a pyroelectric detector that provided a pulse-by-pulse record of the transmitted energy. The backscattered and transmitted signals were measured with peak-reading voltmeters, and the results were displayed on a chart recorder. The data were reduced by taking the ratio of the normalized P(38) signal to the normalized P(20) signal and using the calculations shown in Figure 25 to determine atmospheric temperature. The H_2O content--a value required for use of the Figure 25 calculations--was obtained with a wet-bulb/dry-bulb thermometer.

Lidar data and in situ temperature data from a thermometer near the lidar site were obtained on a number of days from March 14 to October 5, 1979. To provide the most general test of the theory, these measurements were performed over a temperature range as wide as possible.

The results of these measurements are summarized in Figure 26. The error bars represent the combined random error measured from each of the four required signals (the transmitted signal and the backscattered signal at each of the two wavelengths) on a multipulse basis. The average uncertainty in the measurement of T_{rel} is 1.8%, which corresponds to an uncertainty in temperature of approximately 1.1°C . The major contributions to this uncertainty are: pulse-to-pulse variations in laser power, miniarcs in the laser cavity, atmospheric scintillation, detector and other noise from electronic components, variations in aerosol scattering and movement of the target foliage in the wind. Pulse-to-pulse variations in laser power can be the dominant noise mechanism if no provision is made for pulse-to-pulse normalization of the backscattered signal with respect to the transmitted signal. Variations in laser power contribute to the random error in both transmitted and backscattered signals. If

Table 10

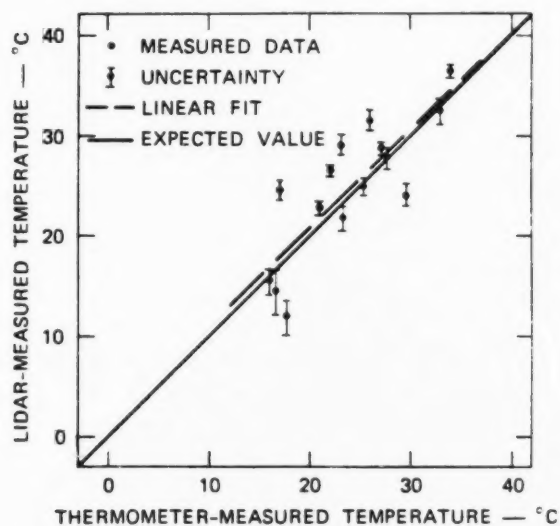
SYSTEM PARAMETERS FOR PATH-AVERAGED MEASUREMENTS OF TEMPERATURE

CO₂ TEA Laser

Transmitted energy	1.5 J
Lines	P(20) and P(38)
Frequency	944.1948 and 927.0091 cm ⁻¹
Pulsewidth	250 ns (full-width half maximum)
Beam divergence	1.0 mrad (to the 10% point)
Typical PRF	0.25 Hz

Receiver

Telescope diameter	31.75 cm
Field of view	2.8 mrad (to the 10% point)
Hg-Cd-Te detector	
D*	$1.1(10^{10}) \frac{\text{cm Hz}^{1/2}}{\text{W}}$
Time constant	0.075 μs
Size	1 x 1 mm

FIGURE 26 SUMMARY OF TEMPERATURE MEASUREMENTS--
INCLUDING LINEAR FIT TO EXPERIMENTAL DATA

pulse-to-pulse variations in laser power are a major contribution to measurement error, analog normalization will eliminate this source of error.

Figure 26 also shows a linear-curve fit to the experimental data. This linear fit has a slope of 0.99 and an intercept of 1.1°C , and thus agrees with the expected values. The correlation coefficient between the lidar-measured temperature and the thermometer-measured temperature was calculated to be 0.825.

One would not expect perfect agreement between the two sets of measurements because the lidar and the thermometer measure different quantities; i.e., the lidar measures the average temperature over the 5-km path, whereas the thermometer measures the temperature at a single location. The water content can also vary over the path, introducing additional discrepancy. Should the error caused by variable H_2O content be unacceptably large, the laser can be tuned to different wavelengths and the average water content along the path measured. In spite of these potential sources of discrepancy, both sensors should record high values on warm days and low values on cool days; to that extent, the lidar and thermometer data are well correlated.

The path-averaged measurements were limited by a random error of approximately 1.1°C . This technique appears useful for making single-ended, path-averaged measurements of temperature. Shorter paths will yield less differential absorption on the wavelengths selected for this study, and it may be necessary to use more strongly absorbed laser lines to maintain accuracy.

B. Stanford University

1. Overview

Byer and Garbuny (1973) theoretically analyzed remote atmospheric pollutant monitoring by absorption. The two methods considered were long-path absorption measurements using topographical targets and depth-resolved absorption measurements using Mie scattering in the atmosphere as a distributed reflector.

Progress in remote monitoring since that first theoretical study has been delayed due to the slow development of tunable laser sources. Depth-resolved absorption measurements, now referred to by the acronym DIAL lidar, have now been demonstrated in the visible, ultraviolet and infrared spectral ranges. The success obtained in using a tunable source lidar for remote measurements has been such that it is presently accepted as the primary means of measurement for making long-range atmospheric measurements using laser sources.

Stanford University's early research work in this area was concentrated on development of a high-power tunable source for remote monitoring applications. Under early NSF support, we developed the concept of, and demonstrated, a tunable infrared source based on a Nd:YAG pumped LiNbO_3 optical parametric oscillator (OPO). The early device demonstrated a tuning range from 1.4 to 4.0 μm with a linewidth of 4 cm^{-1} at an output energy of 10 mJ/pulse at 10 Hz.

The wide infrared tuning range and all-solid-state construction of the tunable source showed its potential use in infrared remote monitoring. The solid state construction offered the potential for reliable operation, and the wide infrared tuning range included the molecular absorption bands of a wide variety of pollutant molecules. An additional benefit of the infrared source is eye-safety operation during either daylight or nighttime.

The OPO tunable source was employed in remote measurements at Stanford in 1977-1978. The Stanford remote monitoring system, which includes a 16-in. diameter receiving telescope, minicomputer and Nd:YAG

laser source was designed and constructed under NSF and EPRI support. The early measurements of SO_2 and CH_4 by Baumgartner and Byer (1978a,b) were conducted as a demonstration of the measurement potential of the system.

Following the early measurements, an extensive program of research was carried out on the OPO source and a parametric amplifier (OPA) [Baumgartner and Byer (1979)] for increased energy. This program led to significant improvements in the tunable source energy, linewidth control, and automatic computer control [Brosman and Byer (1979); Baumgartner and Byer (1979)]. It also led to a design that demonstrated long-term reliable operation under full automatic computer control. The improved OPO/OPA source was used as a transmitter in the system reported here.

Single-ended, remote atmospheric measurements require considerable transmitted energy. For example, measurements using topographical targets require typically 0.01 to 0.1 J/pulse of transmitted energy. DIAL lidar measurements require substantially more energy (0.1 to 1.0 J) due to the weak signal return. Thus, progress in remote monitoring requires increased tunable laser capability.

Stanford University has carried out independent research to investigate new approaches to high pulse energy tunable infrared sources. The results show that there is good potential for development of transmitter sources based on Nd:Glass and on pressure-tunable CO_2 followed by Raman shifting. These future high-energy sources make the potential for depth-resolved infrared lidar appear particularly promising.

The measurement program described in this report illustrates the advantages of lidar for remote monitoring. These advantages include rapid measure time, path-averaged measurements, multiple pollutant capability and excellent measurement precision. The measurement capability will improve as new higher-energy sources become available.

2. Tunable Laser Sources

The requirement for 10 to 100 mJ of tunable infrared laser energy for remote monitoring has led to the development of a continuously-tunable parametric laser source.

The tunable source is part of a remote monitoring system which includes: (1) the tunable laser source, (2) the transmitter and telescope receiver optics, (3) a detector and processing electronics. The system was designed with the goal of allowing real-time remote monitoring measurements to be made by a single operator working from the mini-computer keyboard. Thus the computer controls the tunable source wavelength, the telescope pointing direction, and the data processing and display. The total system has operated as expected and demonstrates that despite its high initial cost, its ease of operation, reliability, and versatility make it an economic method of air pollution monitoring.

a. The Nd:YAG Pump Laser Source

The operation of a parametric oscillator requires a diffraction-limited pump laser source. We previously used a TEM₀₀ mode Nd:YAG oscillator [Byer et al. (1975)] operating with a 780- μ m spot size and 5 to 8 mJ of output energy to pump a Nd:YAG amplifier chain. The amplifier chain, which consisted of two 1/4-in. amplifiers, one 3/8-in. amplifier and two Faraday rotator isolators for stability, provided over 380 mJ of output energy at rates up to 10 pps. The output energy was limited by the small spatial filling factor of the Gaussian beam. To prevent mode clipping and resulting diffraction problems, the beam spot size was chosen at the 1% intensity loss or the $3w_0 = d$ condition, where w_0 is the Gaussian beam spot size and d is the Nd:YAG rod diameter.

To use the Nd:YAG rods more efficiently, we investigated the possibility of using a Nd:YAG unstable resonator oscillator [Herbst et al. (1977)]. Our first design, which took into account the focusing properties of the Nd:YAG rod, used a 1/4-in. dia. rod and an output magnification of 3.3. The resulting output coupling was slightly over 80%. The oscillator operated with up to 200 mJ of output energy in a diffraction-limited beam. Using this source operating at 150 mJ at 10 pps, we were

able to demonstrate 30 mJ of $0.532\text{-}\mu\text{m}$ output by second harmonic generation in Type II KD*P, 10 mJ of $0.3547\text{ }\mu\text{m}$, and 10 mJ of $0.266\text{ }\mu\text{m}$.

Figure 27 shows the Nd:YAG unstable resonator/amplifier system now employed to pump the LiNbO_3 tuner. The available output energy after amplification by the 3/8-in. amplifier is 780 mJ at 10 pps. Thus, the use of the unstable resonator oscillator allows considerable improvement in output energy and a simplification of the Nd:YAG pump system. The gain in energy is largely due to the better geometric filling factor provided by the unstable resonator mode. It should be noted that the mode has a plane wave front and in the near field still has the output mirror zero intensity hole in the beam center. In the far field, the mode transforms to a modified Airy disk intensity function, which can couple very well to Gaussian profile beams. The results cited above were obtained in the near field.

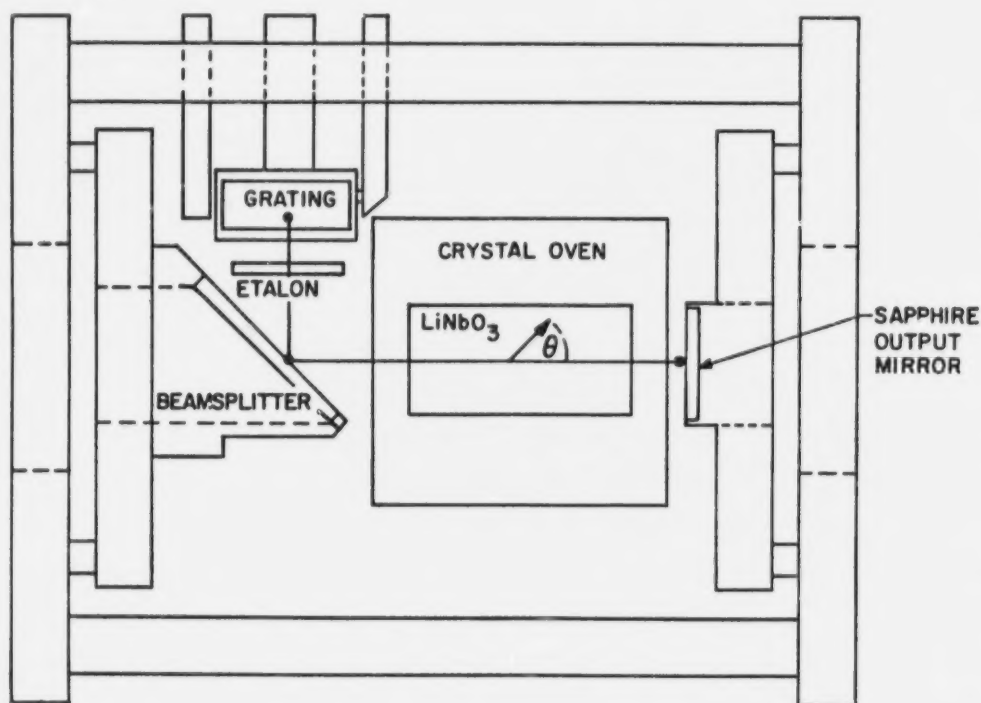


FIGURE 27 SCHEMATIC OF THE LiNbO_3 PARAMETRIC TUNER DESIGN. The $1.06\text{ }\mu\text{m}$ pump radiation is incident from left through the dichroic beamsplitter. A minicomputer controls the stepper motors which scan the grating and crystal angles for wavelength tuning.

The Nd:YAG unstable resonator/amplifier system operates with a 1/2% wall plug efficiency and for up to 1000 h before a flash-lamp change is required. The efficiency and reliability of the Nd:YAG laser source are important parameters in designing a remote monitoring system.

b. LiNbO₃ Parametric Tuner

The LiNbO₃ parametric oscillator operates over the 1.42- to 4.2- μ m tuning range shown in Figure 28 as a singly-resonant oscillator (SRO) with the 1.42- to 2.1- μ m signal wave resonated with a 13-cm long optical cavity formed by a grating and 50% reflecting output coupler. The grating provides linewidth control to less than 4 cm^{-1} . In addition, the grating provides an absolute frequency reference to within $\pm 1 \text{ cm}^{-1}$ over the tuning range of the oscillator.

The parametric tuner is controlled by an interactive program using a PDP-11E10 minicomputer acting through a CAMAC controller. The software allows the setting of the initial wavelength, scan range, scan rate, and repeat scans. It also allows control of the line-narrowing tilted etalon for higher resolution scans. All of the measurement results reported in this section were made with the LiNbO₃ tunable source under automatic computer control.

For 100 mJ of 1.06- μ m input energy, the parametric tuner generates a total of 20 mJ output energy in the signal (1.42 to 2.12 μ m) and idler (2.12 to 4.2 μ m) beams. The device is stable and reliable having operated in a research environment for a 10-month period without degradation of the optical components.

As an example of the control possible with the tunable source for atmospheric measurements, Figure 29 shows an atmospheric absorption spectrum over a 1.5-km path length at a 1.0 cm^{-1} resolution near the CH₄ absorption region; in addition, the figure shows the CH₄ spectrum taken in the laboratory.

This spectrum illustrates the advantages of the continuously-tunable LiNbO₃ parametric source for atmospheric measurements. The wavelength can be chosen to avoid interference from water vapor and to optimize the pollutant absorption cross-section for the measurement.

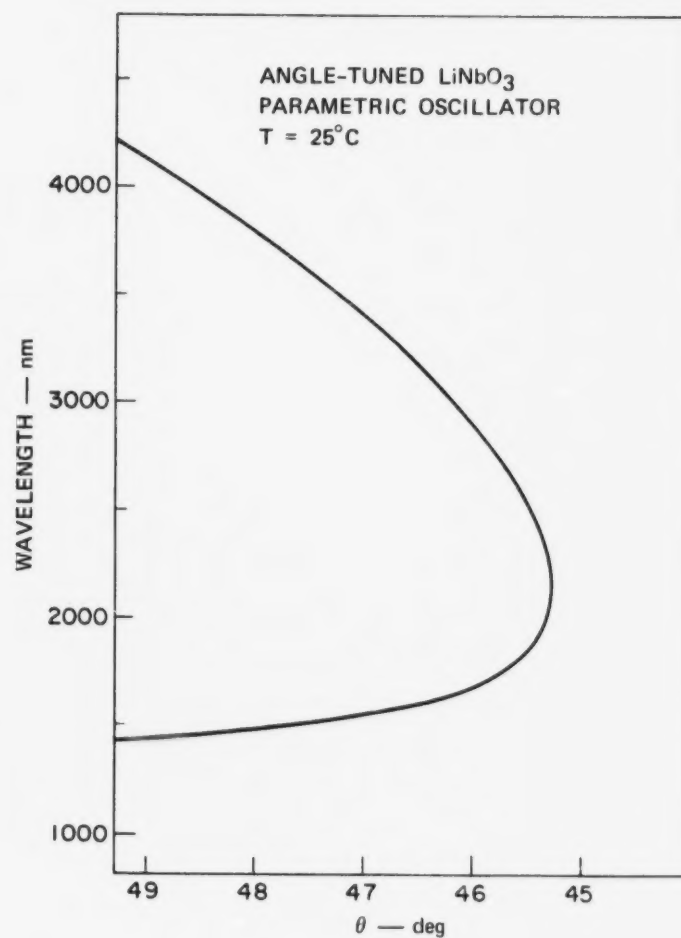


FIGURE 28 TUNING RANGE FOR THE ANGLE-TUNED $1.06\text{-}\mu\text{m}$
PUMPED LiNbO_3 PARAMETRIC OSCILLATOR TUNER

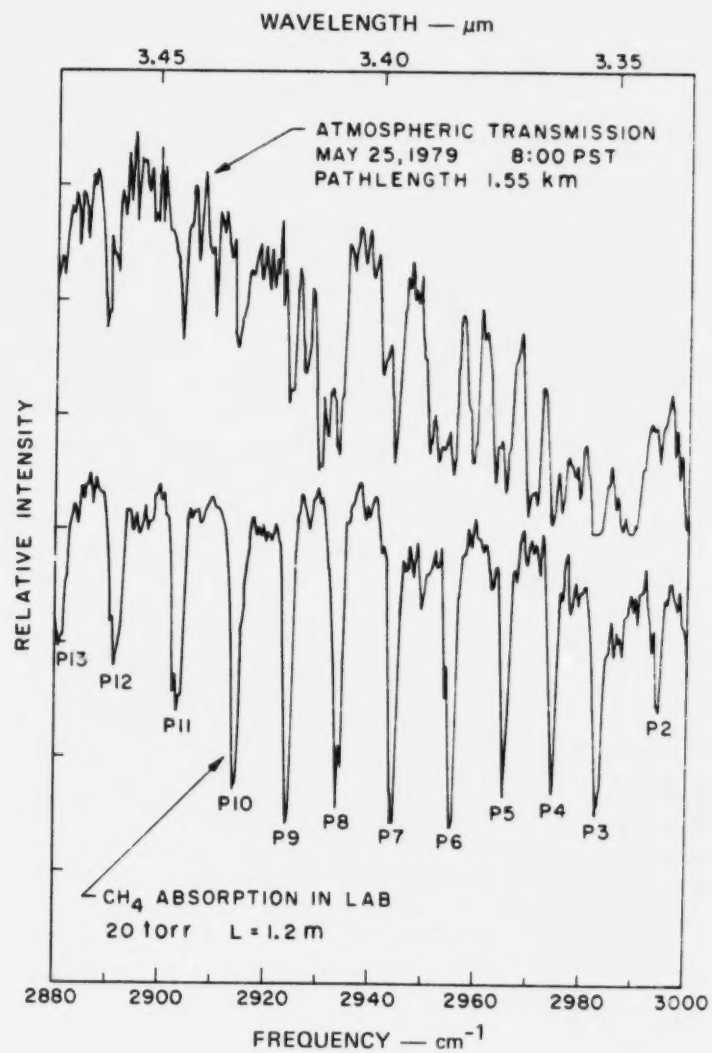


FIGURE 29 A SEGMENT OF AN ATMOSPHERIC ABSORPTION SCAN AND AN IN-LABORATORY ABSORPTION SPECTRUM NEAR THE CH₄ FUNDAMENTAL ABSORPTION BAND AT 3.4 μm

c. Extended Ultraviolet and Visible Tunable Source Based on Nd:YAG

The unstable resonator Nd:YAG source was invented in response to the need for a high-energy tunable source for remote monitoring. Application of the source at Stanford has been to pump the LiNbO_3 tunable infrared source.

However, the Nd:YAG source can be efficiently frequency-doubled and -tripled and used to pump a tunable dye laser source. This source has been extensively developed commercially and is now the tunable source of choice for visible and ultraviolet applications. The use of the unstable resonator Nd:YAG-pumped dye laser for remote monitoring of SO_2 , O_3 , and NO_2 in the ultraviolet was described in subsection a, above.

This laser tool will have widespread impact on a number of fields outside remote monitoring including chemistry, spectroscopy, and biology.

3. Remote Monitoring System

The Stanford remote monitoring system consists of a parametric tuner source, a 16-in. f3-dia. receiving telescope, InSb detector and amplifiers, an analog to digital (A/D) converter, a fast buffer memory, and a PDP-11E10 minicomputer. The system operates in a radar fashion using topographical targets or backscatter from atmospheric particulate matter as retroreflectors. The transmitter operates at 10 pps repetition rate allowing near real-time remote measurements. A single operator working from the minicomputer can control the system including wavelength selection, telescope direction, and data verification displays and recording.

Figure 30 shows a schematic diagram of the optical components of the system. The parametric oscillator supplies 10- to 30-mJ pulses with a bandwidth of approximately 1.0 wavenumber in the wide tuning mode. This bandwidth is adequate to measure concentrations of many pollutants. A detector monitors the transmitted output from the filter for purposes of triggering the received signal recording apparatus and providing an output level reference. To minimize divergence of the transmitted beam, to monitor its range, and eliminate eye hazard, the parametric oscillator

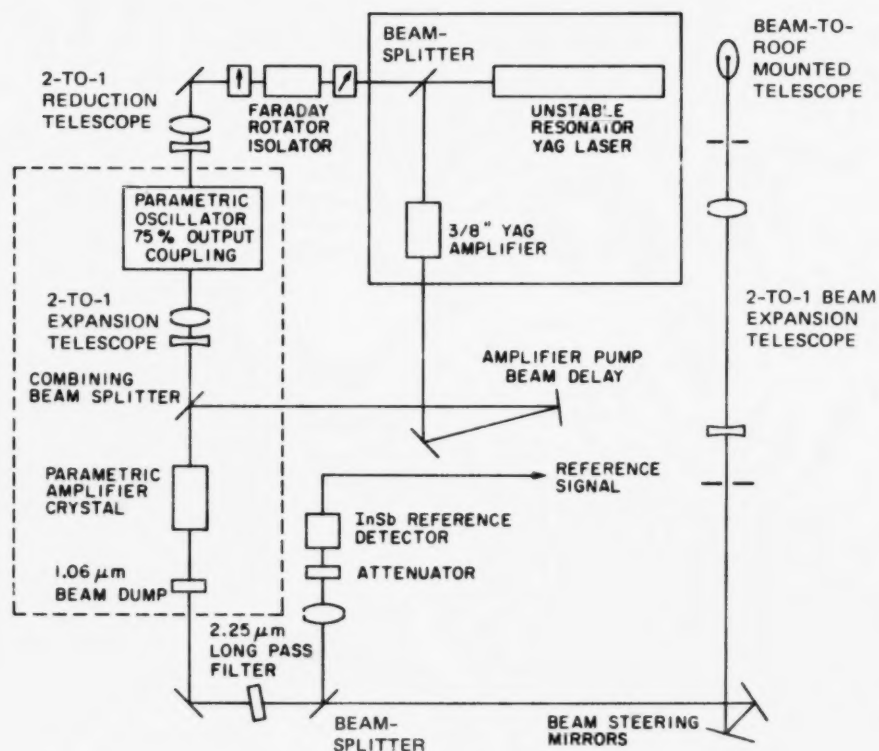


FIGURE 30 SCHEMATIC DIAGRAM OF THE TUNABLE SOURCE MONITORING OPTICS, AND TELESCOPE RECEIVER

beam is expanded to a 2.5-cm radius with a Galilean telescope using calcium fluoride lenses. Finally, by means of various diagonal mirrors, the beam is transmitted from the parametric oscillator to the telescope and into the atmosphere.

The telescope was designed specifically for remote monitoring and uses an alt-azimuth mounting geometry with 45° reflecting mirrors in the x, y axes to reflect the transmitted beam onto an axis collinear with the telescope field of view. To reduce costs, the telescope drive train was designed for ± 10 -min pointing accuracy so that a belt drive and clutch system could be employed. This enables the telescope to be quickly manually positioned without disengaging gears. Two encoders keep track of the pointing direction at all times.

The telescope focuses the received energy onto a 1-mm^2 InSb photo-voltaic detector, which operates at 77 K. The detector size is twice the diffraction-limited focal spot size of the telescope. The field-of-view of the system is 0.41 mrad, which is near the atmospheric limit. Therefore, alignment of the optics is very critical and a method of optical alignment using a helium neon laser was devised. Basically, the procedure involves aligning the components from the source to the detector in sequence. Final adjustments are made by monitoring the return signal. System alignment is stable even though the 750-lb telescope is mounted on an 8-in. dia. aluminum pipe, 30 ft above the laboratory floor. Finally, the telescope was designed to be easily installed in a van for future field use.

The output of the detector is amplified by a preamplifier and a computer-controlled gain-switchable amplifier with an overall bandwidth of 1 mHz. As the return signals arrive at the receiver, they are amplified by the amplifier and digitized in a 5-mHz 12-bit A/D converter and stored in a fast buffer memory prior to processing by the computer. To detect a particular pollutant, the frequency of the transmitted signal may be changed from pulse to pulse. The return signal on the absorption and off the absorption of a particular material may then be examined. At the end of a sequence of pulses the accumulated data are accessed by the PDP-11/10 minicomputer to derive the concentration of the atmospheric constituent of interest.

Figure 31 shows a schematic of the data processing system. Its primary features are a real-time display of the received signal via the buffer memory and direct computer control for data storage and processing.

The buffer memory is particularly useful in this type of application. It serves as a control unit for the digitizing process as well as a storage medium. The primary capability of the unit is that of receiving parallel data from an A/D converter at up to 10 mHz bit rates. The memory contains 1024 16-bit words of bipolar semiconductor random access memory (RAM) along with appropriate control logic. The user can select as many digitizing operations per laser shot as he desires. When the memory is

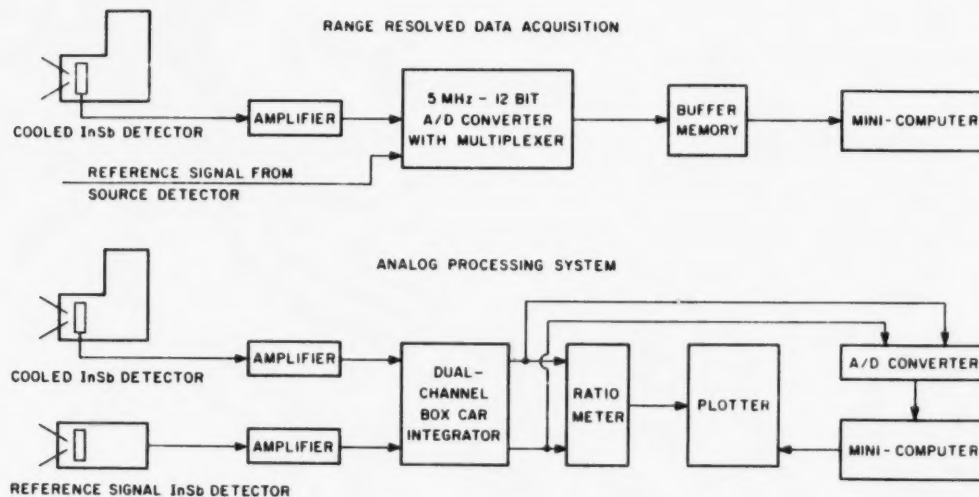


FIGURE 31 SCHEMATIC OF THE RECEIVER ELECTRONICS AND DATA PROCESSING SYSTEM

full or half full, depending on the user's choice, the information it has received can be displayed on an oscilloscope containing two built-in digital-to-analog (D/A) converters. In addition, an interrupt signal is generated which tells the PDP-11/10 computer system that it can access the data that have been accumulated. Because of the Unibus type of architecture of the PDP system, the buffer memory can be treated as an additional 1 K of memory. Since the output of the A/D is 12 bits, the other 4 bits in each word can be used to contain housekeeping data, such as various amplifier gain settings. In addition, at the conclusion of the digitization of the A/D data, additional memory positions are reserved for more housekeeping data, (i.e., transmitter output level), that may also be digitized by selecting other inputs of the multiplexer in front of the A/D.

Figure 32 shows an example of a return signal. The first peak in the return is due to backscattering from particulate matter in the atmosphere. The second and larger peak is from a stationary target, in this case, Hoover tower, located at a distance of 775 m from the telescope. For this particular return, the transmitter wavelength was 1.06 μm and the transmitted power was 12.5 mJ/pulse. The received power from the stationary object return is of the order of 1 mW for a transmitter power of about 1 MW.

During the past year, a series of improvements have been made in the monitoring system in both the hardware and the software. These improvements have led to a reliable, fully automated, remote measurement system which has resulted in long-term measurements of CH_4 and, more recently, H_2O .

Figure 33 is a schematic of the present system. To prevent damage to the LiNbO_3 crystals in OPO and OPA, two important changes were made. First, the cavity length of the unstable resonator Nd:YAG laser, which is the pump source for the OPO and OPA, was doubled to 1.50 m to increase the pulse length to 18 ns and therefore to decrease the peak optical intensity. This avoids the danger of crystal tracking due to self-focusing effects. The second change was the introduction of a far-field converter, which transformed the doughnut-shaped near-field laser output beam into an Airy-disk profile, which ensures better beam homogeneity. This in turn lowers the threshold of the OPO and also avoids hot spots in the beam profile and thus reduces the chance of crystal surface damage.

About 1% of the tunable output is reflected into a photo-acoustic cell to continuously monitor the cross section of the observed species. An absorption cell is used to measure the absolute absorption cross section of a pollutant at a given wavelength.

Figure 34 shows a schematic of the improved data processing and control electronics. The signals from the cooled InSb detector and the reference detector are integrated over one shot in a gated integrator and read out by the computer which ratios and averages them. In one mode the computer tunes the OPO on and off a resonance line, uses the signal from

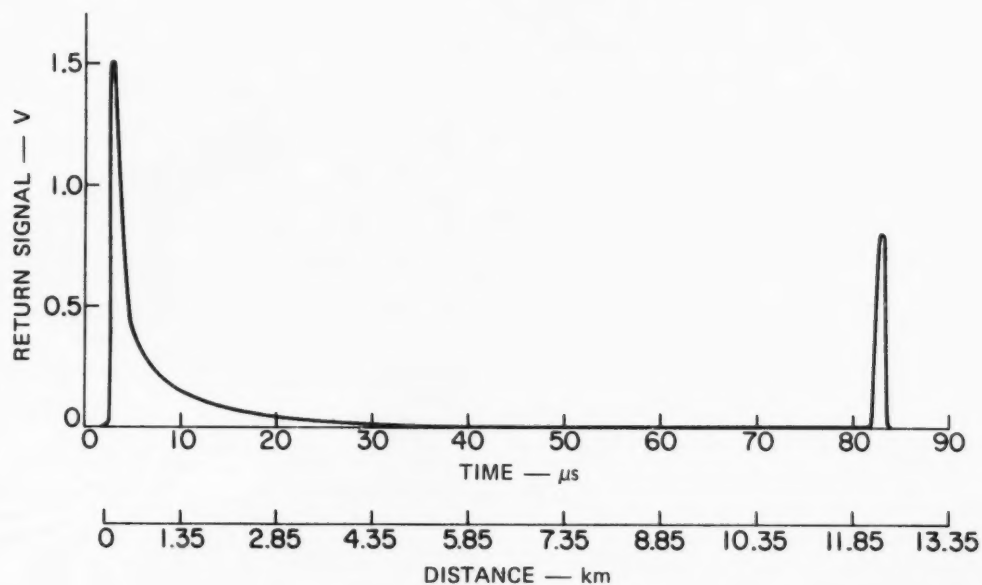


FIGURE 32 EXAMPLE OF A LIDAR RETURN SHOWING BOTH MIE BACKSCATTER AND TOPOGRAPHIC RETURNS WITH $\lambda_t = 1.06 \mu\text{m}$ AND $E_t = 45 \text{ mJ/PULSE}$

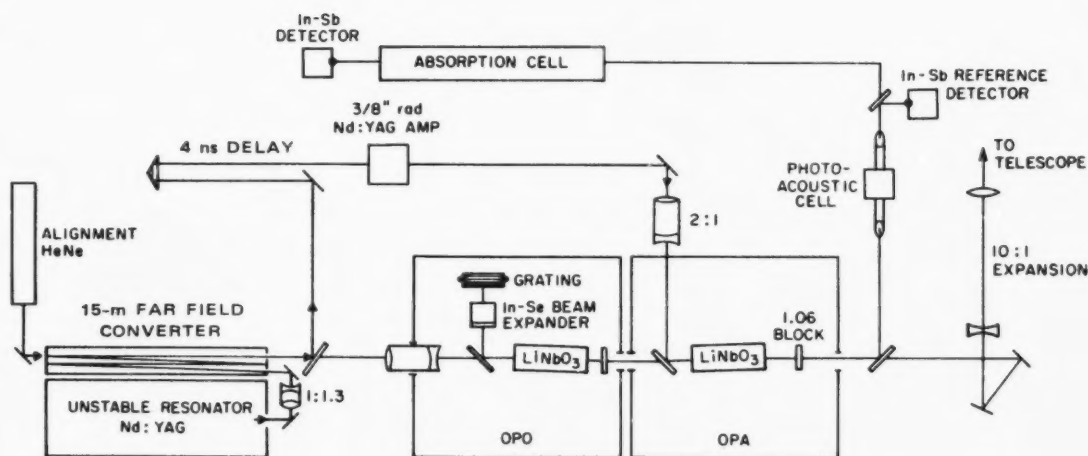


FIGURE 33 SCHEMATIC OF THE Nd:YAG LASER PUMPED LiNbO_3 TUNABLE SOURCE TRANSMITTER. The source operates at 10 pps with 20 mJ output energy tunable under computer control over a 1.4 to 4.0 μm range at 1.0 cm^{-1} linewidth. An etalon can be used to narrow the linewidth to less than 0.1 cm^{-1} .

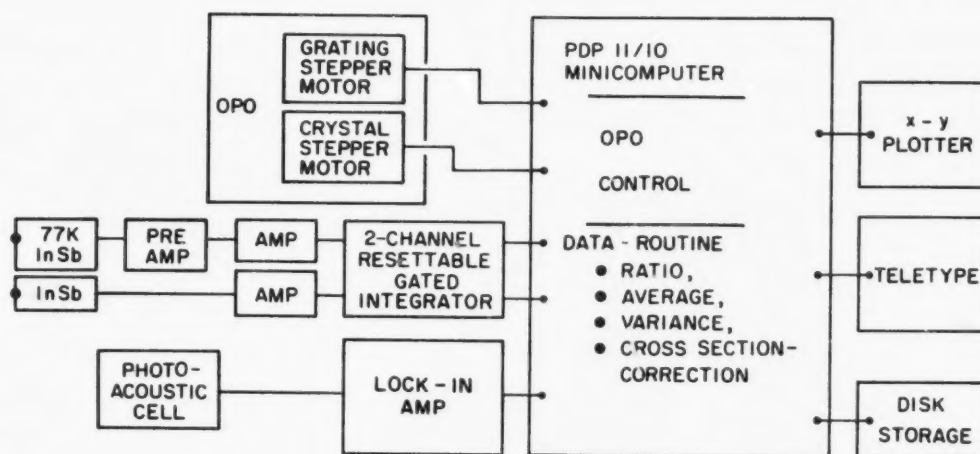


FIGURE 34 SCHEMATIC OF THE ELECTRONIC CONTROL AND DATA PROCESSING SYSTEM BASED ON THE PDP-11 MINICOMPUTER

the photoacoustic cell to correct for slow changes of the absorption cross section, and prints immediately the measured concentration or stores the data for laboratory processing. Another operating mode provides automatic atmospheric or in-laboratory transmittance scans over wide spectral regions. The control program simplifies the handling of the system considerably.

During this program important improvements were made to the remote monitoring system. The tunable source was modified by adding a prism beam expander to the OPO resonator cavity. This led to a reduction in the grating-only linewidth from 4 cm^{-1} to 0.9 cm^{-1} . The OPO/OPA system was enclosed in a dust-free housing and operated at a lower energy level to ensure long-term, damage-free operation. Reliable operation from January 1979 to November 1979 verified that the all-solid-state tunable source can indeed be trouble-free for extended operation times.

A tilted etalon for linewidth control to 0.1 cm^{-1} was installed. The galvanometer tilt control proved to have inadequate resettability and is being replaced by a stepper motor device. However, the etalon did operate over an extended 40 cm^{-1} scan range under computer control. When the new mechanical system is completed we expect to conduct atmospheric measurements at the improved resolution.

The second area of improvement was the system control software. The software package now allows control of the tunable source, monitoring of an absorption cross section via the photo-acoustic cell, normalization of the transmitted pulse energy, data collection and processing of the returned signal. Software was also developed to display the recorded spectrum in real time or to calculate and plot the measured pollutant concentration.

4. Measurements of SO₂, CH₄, and H₂O Using the Stanford Lidar System

The measurement program at Stanford was designed to complement the measurements made at SRI. Thus, the Stanford measurements emphasized continuous tunability and its advantages. Measurements made of SO₂ at 4.0 μm , of CH₄ at 3.3 and 1.66 μm , and of H₂O at 1.7 μm were used to demonstrate this capability.

A second goal of the Stanford measurement program was the demonstration of fully automated measurements over an extended time period. To this end measurements were made of CH₄ over an 18-h period. The results were compared with those from a nearby point monitoring station. Future measurements are planned for H₂O, atmospheric temperature, CO, ethane, and CO₂. These measurements require the use of the tunable source in a narrow linewidth mode. Consequently, improvements in the line-narrowing device are being made and measurements are expected to begin in the near future.

a. SO₂ Measurements

Because of its importance as an atmospheric pollutant, SO₂ was chosen as the demonstration species in the measurement program. The results of these measurements at 4.0- μm wavelength were published by Baumgartner and Byer (1978a) and are summarized here.

Measurements of SO₂ vapor with a remotely-located sample chamber allowed direct comparison of lidar measurements with known gas concentration values. The lidar was also tested over a wide range of gas concentration values and the system sensitivity was determined using the sample chamber.

The 4.0- μm spectral region is advantageous for monitoring SO_2 for three reasons: the effective absorption coefficient value for the $\nu_1 + \nu_3$ combination band is only a factor of 10 smaller than the resonances beyond 7.0 μm ; the daylight operation difficulties of an ultra-violet or visible Raman measurement are avoided; and interference problems from other pollutant bands are almost non-existent at 4.0 μm as shown by Haught and Dowling (1977). From the basic lidar equation,

$$c = \frac{1}{2(k_{\text{on}} - k_{\text{off}})L} \ln \frac{T_{\text{off}}}{T_{\text{on}}}$$

the relation is easily derived where c is the concentration in atmospheres, $k_{\text{on}} - k_{\text{off}}$ is the absorption coefficient in $(\text{cm atm})^{-1}$, L is the length of the sample chamber in cm, T_{off} is the round-trip transmission of the lidar off-resonance, and T_{on} is the round-trip transmission of the lidar on-resonance. The value of $k_{\text{on}} - k_{\text{off}}$ was determined from two independent measurements. From Pine and Moulton (1977) a value of $k_{\text{on}} - k_{\text{off}} = 0.372 (\text{cm atm})^{-1}$ was obtained; k_{on} corresponds to a 4-cm^{-1} average bandwidth centered at 2509.5 cm^{-1} and k_{off} to a 3-cm^{-1} average bandwidth centered at 2501 cm^{-1} . A laboratory scan with a 4.0-cm^{-1} bandwidth parametric source and the same analysis as above yielded a value of $k_{\text{on}} - k_{\text{off}} = 0.363 (\text{cm atm})^{-1}$. A White cell of 675-cm pathlength with SO_2 of 1 Torr in atm of N_2 was used in the laboratory measurements.

Following determination of the absorption coefficient, the telescope was aligned to make measurements through the remotely-located sample chamber. Two sample chamber scans are shown in Figure 35. The lidar transmitted energy was 0.6 mJ/pulse. Each curve is the average of three or four sequential frequency scans taken to reduce the variance. Standard deviation error bars are the result of the averaging at each source center frequency point. The darkened regions are frequencies that were combined to obtain SO_2 concentration values. The resultant averaging effectively uses 500 laser shots for each gas concentration data point. The lidar measurement of SO_2 requires a two-step process. The chamber is purged of the gas to be measured, and a reference scan is taken to obtain a 100% transmission as a check on the system stability. The gas is

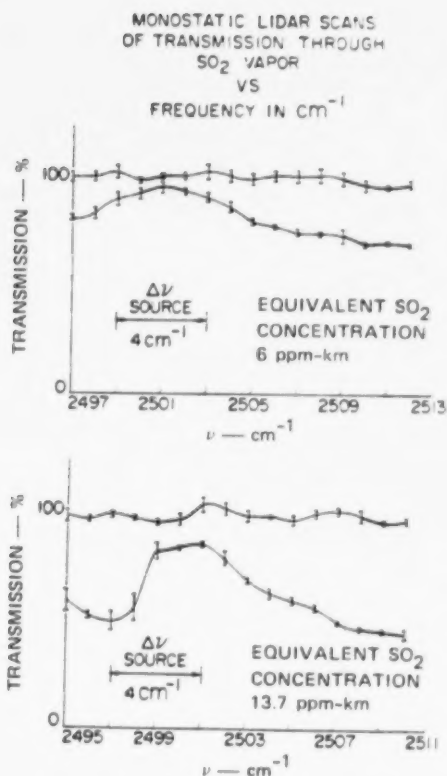


FIGURE 35 EXAMPLES OF LIDAR FREQUENCY SCANS THROUGH A REMOTELY-LOCATED SAMPLE CHAMBER FOR TWO VALUES OF SO₂ CONCENTRATION WITH NORMALIZATION SCANS

then transferred from a prechamber of known volume, temperature, and pressure and mixed with sample chamber air by a stirring fan. Finally, a second set of scans is taken to determine gas concentration with the lidar. The results of measurements for various concentration values, with the appropriate measurement uncertainties, are displayed in Figure 36. The error bars represent standard deviation errors. The horizontal error bars result from a 3% uncertainty in gas concentration, while the vertical bars are indicative of measurement uncertainty in the lidar measurement. The sensitivity of the lidar for SO₂ measurements is

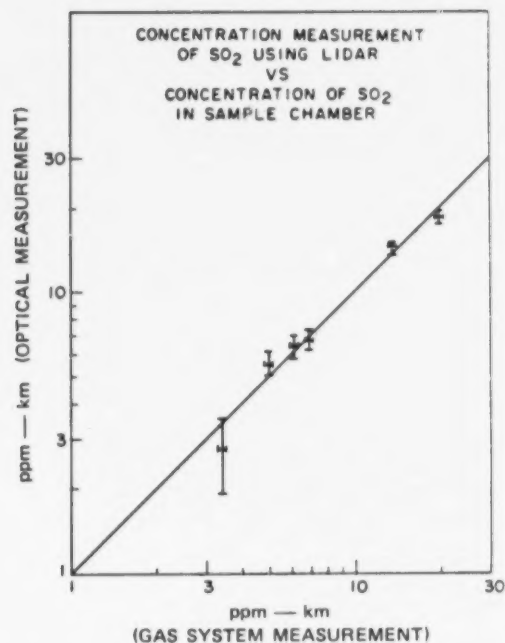


FIGURE 36 A COMPARISON OF LIDAR MEASUREMENTS WITH IN SITU GAS CONCENTRATION FOR THE REMOTELY-LOCATED SAMPLE CHAMBER

0.9 ppm-km, which is derived by extrapolating the measurement to a unity SNR. The linewidth value of 4.0 cm^{-1} for this measurement is appropriate since the SO_2 band linewidth is pressure-broadened by the atmosphere to at least that order.

Sample chamber tests were conducted for SO_2 measurements over a wide range of concentration values. The agreement between gas concentration values in the chamber and lidar measurements was within experimental error.

Recent advances in parametric oscillator performance, both with regard to increases in output energy and substantial reduction in linewidth, have been demonstrated. Also, pulsed dye laser oscillator and amplifier combinations for use in infrared generation by mixing have improved substantially [Byer and Herbst (1978)]. Either of these sources driving a

parametric amplifier should provide as much as 40 mJ in both the signal and idler parametric waves, making possible an important improvement in the measurement capability of the infrared tunable system. From the measurement described here, it is possible to project the sensitivity resulting from improved parameter choices. For example, a 10-mJ pulse averaged over 200 s yields a projected SO_2 measurement sensitivity of 0.28 ppm.

For SO_2 , the present sensitivity is sufficient to measure source emissions that are typically greater than 100 ppm. With the projected improvement in sensitivity, background concentrations of SO_2 should also be measurable.

b. Methane (CH_4) Measurements

The CH_4 measurements were conducted in two parts. Early measurements, by Baumgartner and Byer (1978b), made at a 4.0 cm^{-1} resolution at both the overtone and fundamental absorption bands centered at $1.66 \text{ }\mu\text{m}$ and $3.3 \text{ }\mu\text{m}$ were successful.

The purpose of the early measurements was to demonstrate the use of the tunable source for cross-section optimization for short-path or long-path measurements. Thus, measurements at $3.3 \text{ }\mu\text{m}$ are useful for sensitive CH_4 determination over relatively short path lengths of the order of 1 km. Measurements at $1.66 \text{ }\mu\text{m}$, on the other hand, are useful for a long-path ($> 5 \text{ km}$) or for high-concentration CH_4 measurements, such as for a liquid natural gas spill or high-pressure gas line leak.

An interesting by-product of the early measurements was the use of the overtone absorption lines to measure CH_4 in a coal mine. This problem was pursued under a consulting agreement with a major steel company. The results were very encouraging and may yield a laser-based remote CH_4 monitor for use in coal mines.

During the CH_4 measurements, absorption scans of the atmosphere were made to determine the possible interference due to water vapor.

Figure 37 illustrates the tunable source wavelength scanning capability, with Hoover tower, located 750 m from the telescope, serving as

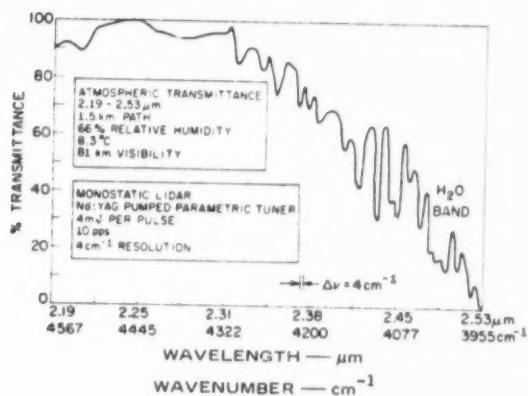
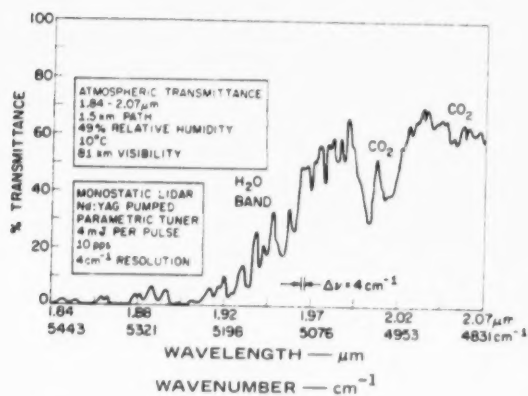
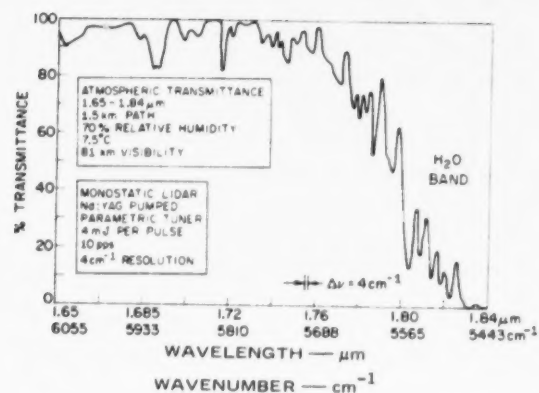


FIGURE 37 ATMOSPHERIC TRANSMISSION SCANS
OVER A 1.5-km PATHLENGTH

the topographic target. Since the wavelength region covered in the scan was quite broad, 100% scans were taken by reflecting some of the output directly into the receiver. The SNR for the scan was 60. Each of the three segments in Figure 37 was scanned in a continuous manner in approximately 10 min. This is a capability similar to a Fourier transform spectrometer system but with a lower resolution. The atmospheric windows within the source tuning range of 1.4-4.0 μm contain absorption bands of many materials that present numerous measurement possibilities. A partial list includes CO, HF, NH_3 , NO_2 , HCl , H_2S , SO_2 , CH_4 , C_2H_2 , C_2H_6 , freons, and, of course, CO_2 and H_2O . In the second scan in the region near 2 μm , there are three combination bands of CO_2 . The term values are 20^0_1 , 12^0_1 , and 04^0_1 for the 1.96- μm , 2.01- μm , and 2.06- μm bands. The two prominent bands at 2.01 μm and 2.06 μm are of interest because of atmospheric temperature measurement possibilities.

Extended single-ended atmospheric absorption measurements of methane were made with the improved tunable source. The typical path length was 1.541 cm, with the Hoover tower as the topographical target. Spectral scans over a wavelength range from 1.45 to 4.0 μm were made.

Reduction of the tunable source linewidth from 4.0 cm^{-1} to 1.0 cm^{-1} led to the significantly better measurement of atmospheric absorption spectra. Figure 38 shows a section of a spectral scan in the 5500 - 5800 cm^{-1} range. The 300 cm^{-1} scan is a part of a scan that extends to 2500 cm^{-1} . The region shown in Figure 38 covers the overtone absorption band of H_2O . The absorption peaks are not noise but are indeed individual absorption lines of water vapor. To illustrate the resolution of the source, Figure 38 shows a section of water vapor absorption over a 40 cm^{-1} interval. In this scan, the water vapor lines have been labeled. The scan has a resolution close to 1 cm^{-1} and, therefore, does not resolve the water vapor lines which have half-widths approximately in the range between 0.03 and 0.1 cm^{-1} . However, water vapor density can be determined with the 1 cm^{-1} resolution if an effective absorption cross section is used.

Figure 29 shows a segment of the atmospheric absorption spectrum near the methane 3.4- μm fundamental band. Both CH_4 and H_2O absorption

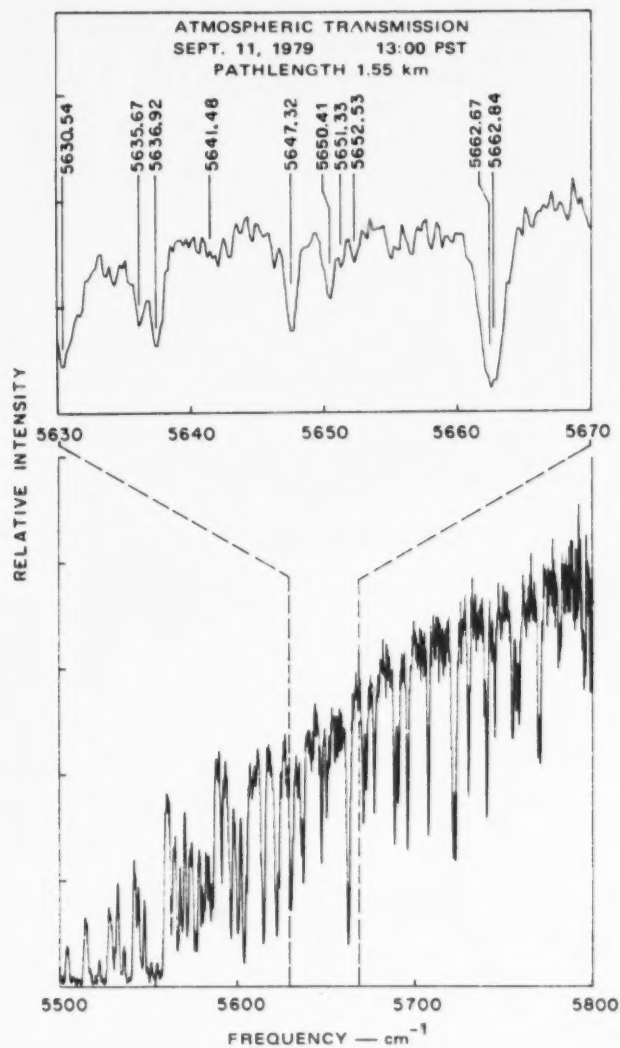


FIGURE 38 A SEGMENT OF AN ATMOSPHERIC TRANSMISSION SCAN OVER A 1.55-km PATH IN THE SPECTRAL RANGE FROM 5500 TO 5800 cm^{-1} (lower figure). The upper figure is a detailed segment of the scan. Here a region of H_2O absorption is shown in which H_2O lines have been identified and labeled.

lines are present. To clearly identify the CH_4 lines and to aid in selecting a transition that is free of water vapor, an absorption spectrum of CH_4 was measured in the laboratory. A section of the scan centered on the P-branch is also shown in Figure 29. Figures 29a and 29b can be overlaid to show that the P(10) methane transition is of proper strength and nearly water-vapor-free for atmospheric measurement over the 1.5 km path.

The ability to select interference-free absorption bands and to verify the location of spectral lines by scanning the spectrum is a major advantage of the tunable source.

We selected the P(10) transition for CH_4 measurements. The computer was programmed to tune onto the P(10) line and off the line every 20 pulses. The data was normalized and processed to show ppm CH_4 . An effective cross section for CH_4 (P10) was used based on the resolution of the OPO/OPA source. To correct for any long-term drift, the cross section was measured continuously during a run by a photo-acoustic detector cell. The value of the cross section was used to calculate the methane concentration in near real time.

It should be emphasized that the present software program is run on a small PDP-11 minicomputer with only 16 K of memory. To operate the program use has been made of program overlay techniques. It should also be stated that the extensive software development has been done over a 6-month period and that it represents a major investment in the system's capability.

Figure 39 shows a segment of an 18-h CH_4 measurement run. Here, the raw measurements are shown for 20 shots/line averaging or 4 s per point. The rms fluctuations are calculated to be 0.6 ppm. To improve the precision of the measurement, longer averaging times can be used. Figure 40 shows a section of the same CH_4 measurement run with an 80-s averaging time. Here, the rms deviation is reduced to 0.03 ppm, which is less than the CH_4 fluctuation level in the atmosphere.

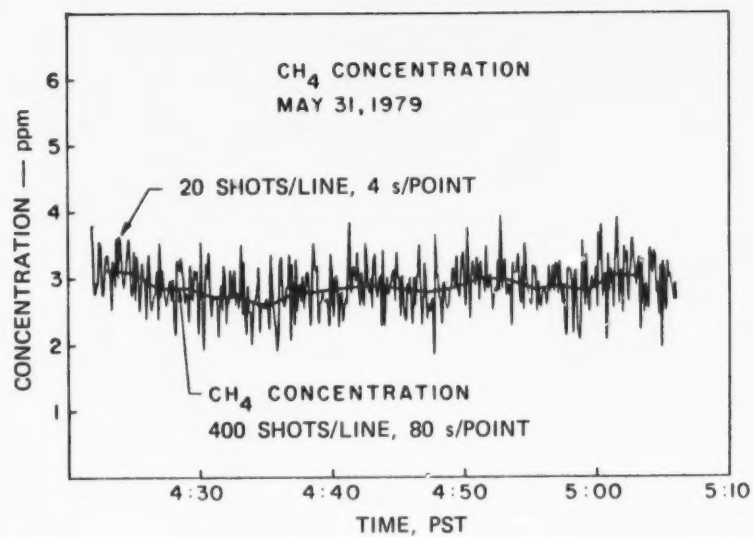


FIGURE 39 A SECTION OF AN 18-h MEASUREMENT OF CH_4 ON MAY 31st, 1979 OVER A 1.55-km PATH. The 20-shot average yields a 4-s measurement time and a 0.6-ppm rms error.

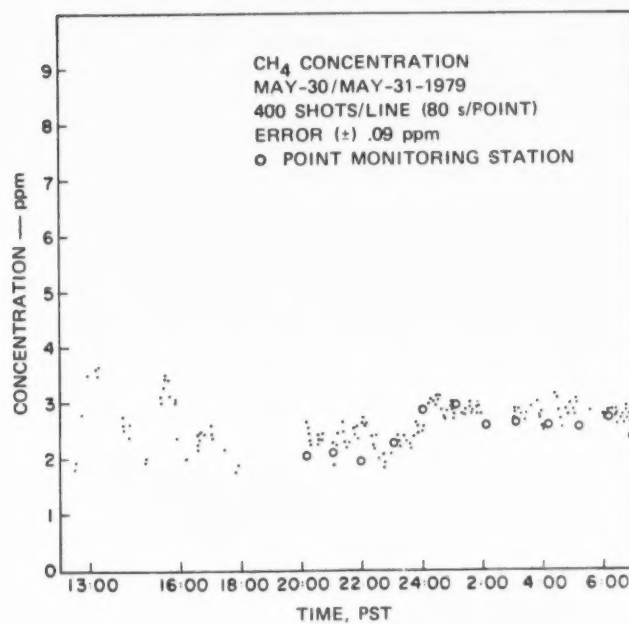


FIGURE 40 A PART OF THE CH_4 MEASUREMENT SHOWING CH_4 FLUCTUATIONS OVER THE PATH BEING MONITORED. The statistical rms noise of the measurement is 0.03 ppm, which is less than the CH_4 fluctuation level.

During these measurements concern developed that the CH_4 measured levels were high since the global average CH_4 level is close to 1.5 ppm. Therefore, hourly CH_4 average measurements were obtained from the Redwood City Bay Area Pollution Control District point monitoring station.

To check further on the correlation between the Stanford measurements and the Redwood City station measurements, a second CH_4 lidar run was made on June 6 and 7, 1979. Figure 41 shows the results. In this case, the wind was blowing down the bay from Redwood City toward Stanford. The wind velocity and distance gave a 20- to 30-min delay time from Redwood City to Stanford. This delay time correlates with the peak CH_4 measurement at 2200 at Redwood City and at Stanford at 2230. Later in the evening when the wind ceased, Stanford measurements became independent of the Redwood City station measurements.

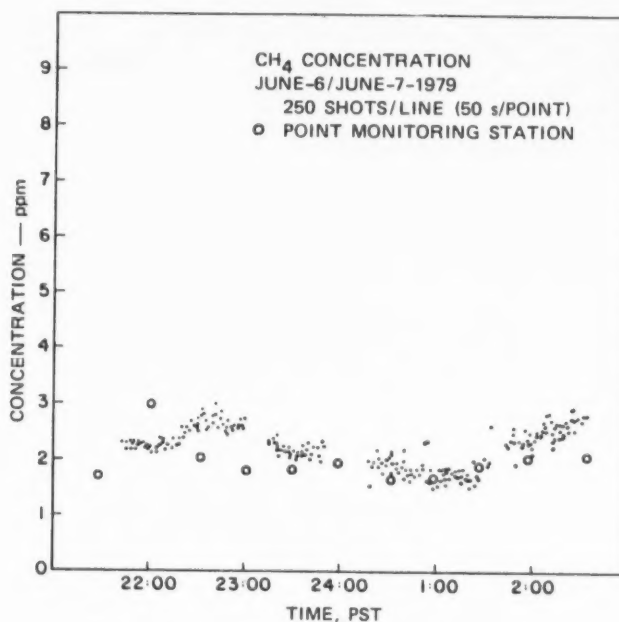


FIGURE 41 CH_4 ppm AS A FUNCTION OF TIME FOR A SECOND MEASUREMENT RUN. The Stanford lidar data are plotted as dots. Here a prevailing wind yielded a 30-min delay between the atmosphere at Redwood City point monitoring station and its arrival near Stanford campus.

It should be noted that the Stanford lidar system was making CH_4 measurements every 50 s. The Redwood City flame ionization detector measurements are averaged over 1-h intervals for recording. Even though the measurement systems are separated physically, the agreement in measured CH_4 levels is excellent as would be expected in a region free of large sources of CH_4 as is the area between Stanford and Redwood City.

d. Water Vapor and Temperature Measurements

During the final phase of this lidar program, we began an investigation of H_2O spectrum for density and temperature measurements. Figure 42 shows a section of the H_2O absorption spectrum near 5650 cm^{-1} . The labeled peaks have been selected for atmospheric temperature measurements due to their proximity in wave number and their widely-spaced lower levels. Here, E_0 denotes the energy above the H_2O ground state in cm^{-1} .

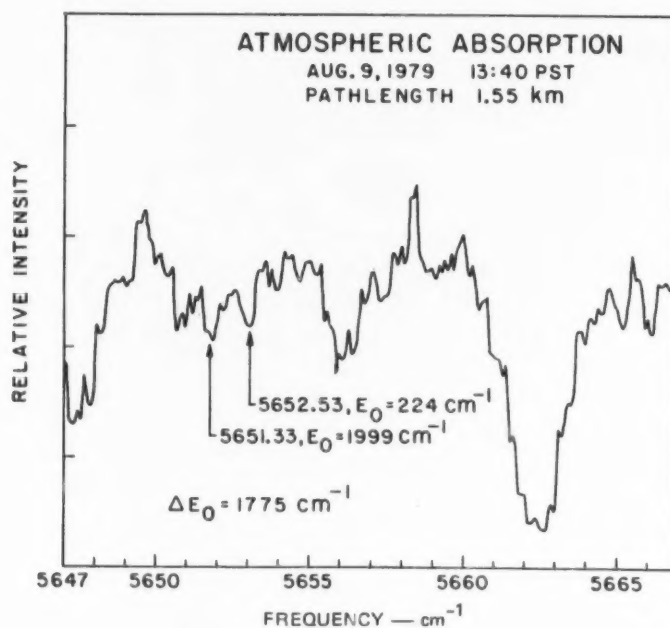


FIGURE 42 A SECTION OF THE H_2O ATMOSPHERIC ABSORPTION SPECTRUM OF USE FOR H_2O AND ATMOSPHERIC TEMPERATURE MEASUREMENTS

A straightforward calculation shows that these two H_2O lines should allow temperature measurements to be made to better than $\pm 1^\circ\text{C}$ for a 1% precision in absorption measurement. Standard CH_4 measurements show that such a measurement precision is possible for an averaging time of approximately 5 min. Preliminary measurements have shown that the peak ratios of the two H_2O lines do vary with atmospheric temperature as expected. Improvements are being made to the linewidth control of the OPO before long-term atmospheric temperature measurements are attempted.

The primary emphasis in these measurements was to demonstrate long-term reliable remote air pollution measurement capability. Efforts have been concentrated on CH_4 rather than to attempt measurements on a wider variety of molecules. However, it should be noted that within the OPO 1.4- to 4.0- μm tuning range are absorption bands of a number of hydrocarbons, including ethane, propane, ethylene, and formaldehyde. Other molecules of potential interest include HCl , HF , N_2O , NO_2 , H_2S , NH_3 , CO , and CO_2 .

5. Investigation of Laser Tomography as a New Wide-Area Measurement Technique

This research program has emphasized the use of tunable lasers for remote air pollution measurements with the long-path absorption and DIAL measurement approach. These remote measurement methods offer the advantages of being single ended and sensitive, and, in the case of DIAL, provide depth resolution. However, the single-ended measurement methods require high transmitted laser power and are limited in range to less than 10 km due to an inverse range squared returned signal dependence.

During the final year of this program Stanford investigated and proposed a new measurement technique for wide area, two-dimensionally, depth-resolved air pollution measurements. The technique is analogous to X-ray tomography and is called laser tomography [Byer and Shepp (1979)]. This technique has potential advantages as a remote measurement tool.

Figure 43 shows a schematic of the laser tomography measurement technique. A central laser source is rotated and illuminates sequentially M_1

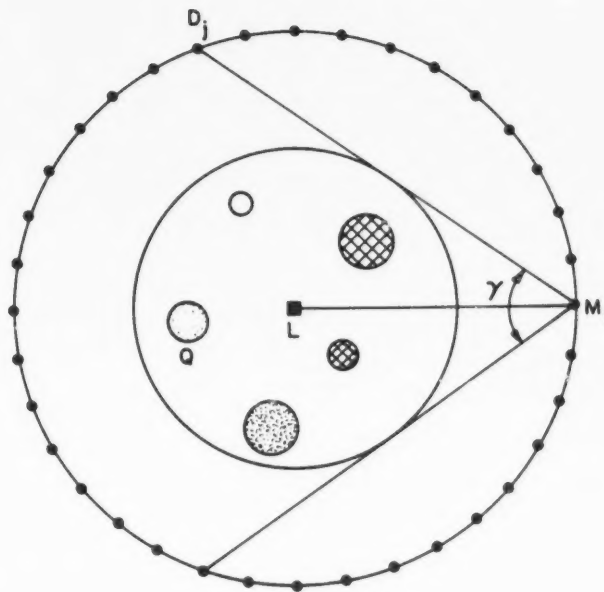


FIGURE 43 SCHEMATIC OF A COMPUTERIZED AIR POLLUTION TOMOGRAPHY MEASUREMENT USING FAN BEAM GEOMETRY. The laser source at L rotates and illuminates mirrors at M_i which act as virtual sources and in turn illuminate detectors D_j . Monitored pollutant sources Q are located within an area A determined by the fan spread angle γ .

mirrors, which fan out the laser beam to illuminate D_j detectors. The absorption across the circle from M_i to D_j is measured and the process is repeated for each M_i . All the absorption integrals are stored and used as input to a computer routine to reconstruct a two-dimensional image of pollutant concentration.

The obvious disadvantage of laser tomography is the need for a monitoring system of mirrors and detectors; typically, 64 to 128 detectors would be needed. However, the disadvantages of setting up the monitor array are offset by the unique advantages of laser tomography. In

addition to two-dimensional pollution concentration measurements, these advantages include low required laser power and wide-area monitoring for distances as great as 50 km.

Computer modeling studies of the details of laser tomography in the past year at Stanford have verified that laser tomography is potentially a very useful measurement approach for wide-area measurements. For example, a 20-km dia. area can be monitored with only 25 W of CW CO₂ laser power. Research on furthering this new measurement technique is being pursued.

III UTILIZATION OF THIS RESEARCH

A. Applications

The development of new remote sensing instruments may be applied to: enforcement of air pollution regulations, testing of air pollution plume models, investigation of atmospheric chemistry of air pollutants, oil and gas exploration via sensing of light hydrocarbons, detection of pipeline leaks, detection and identification of gases of military importance, and exploration for geothermal regions. Instrumentation for these applications may take the form of ground-mobile, airborne, or spaceborne lidar systems.

SRI and Stanford University have vigorously pursued these applications under a number of government and commercial contracts which have resulted from the research conducted under this grant. Publications and presentations relevant to this work are given in the Appendix to this report. Laser devices developed under this grant are now being marketed by commercial organizations.

B. Overall Status of User Interest

The work completed by SRI and Stanford University on remote measurement of air pollutants has generated a large amount of interest among potential users. Additional funds have been received from several organizations to supplement the research program, to apply its early results, and to investigate applications other than measurement of air pollutants. Negotiations are now under way with several prospective users, and many others have expressed interest. These developments assure that the results being obtained are gaining wide acceptance among present and potential users, and that additional users will be found. Details are provided below.

C. Funding Related to Accomplishments

Early research under this grant on visible ultraviolet DIAL has already reached the field applications stage under contract with EPRI. SRI has constructed a self-contained, ground-mobile DIAL system for measurement of NO_2 . This contract, initiated in February, 1972, was funded at the \$520,000 level. This system was successfully tested, and good agreement was obtained between measurements with this lidar and accepted point monitors for both sample chamber and ambient concentrations. Higher laser beam quality and stability is required to generate the ultraviolet wavelengths required to measure SO_2 and O_3 than was available with the transverse flow dye laser. As a result EPRI has awarded a \$530,000 contract to design, construct, and test a second-generation DIAL system using Nd:YAG pumped dye lasers. These lasers have the required beam characteristics. The system will be completed and ready for participation in EPRI's plume model validation program in April, 1980.

Research on infrared DIAL remote-measurement systems initiated under this grant has led to an SRI contract with the U.S. Geological Survey entitled, "Remote Detection of Methane Using a Differential-Absorption Lidar System." The purpose of this work was to apply infrared DIAL to petroleum exploration. The contract began in May 1975 at a funding level of \$19,370. Successful use of a DIAL system was demonstrated. A contract for \$250,000 was awarded by a major oil company for research in the same area. Additional funding is expected.

SRI received a contract from Edgewood Arsenal, U.S. Army Research and Development Services, entitled "Optical Bases for Remote Biological Aerosol Detection," for \$100,000 in December of 1975. The work is directed toward evaluating the fluorescence lidar technique for the remote detection and identification of biological aerosols and uses some lidar equipment and much expertise developed on this NSF-RANN program; \$440,000 has been recently awarded as a follow-on for construction of a ground-mobile system.

During 1976, SRI was awarded two contracts for a total of \$150,000 by the U.S. Air Force to study the feasibility of using optical techniques for remote detection and identification of gases of interest. This contract is a partial outgrowth of results achieved under NSF-RANN sponsorship.

Stanford University has received three contracts to expand on the work being supported by NSF-RANN. NASA-Langley Research Center awarded a research grant of \$40,000 for "Remote Air Pollution Measurements" to augment NSF-RANN funding (February 1974 to February 1975). ERDA gave two research grants for a "Tunable Coherent Spectrometer." One grant (February 1974) was for \$80,000; the other (June 1975 to June 1976) was for \$150,000. The purpose of the research was to develop an analytical tool for isotope spectroscopy and separation. Two spectrometers--copies of the one at Stanford University--have been constructed by ERDA at the Los Alamos Scientific Laboratory.

The Electric Power Research Institute (EPRI) awarded a research contract, "Air Pollution Demonstration Measurement Program," to Stanford University to measure pollutants from a nearby heating plant using the Stanford tunable laser. The initial contract which ran from April 1975 to April 1976 had a value of \$86,000. A renewal of the original contract for an additional year of research at a level of \$98,000 has recently been awarded.

SRI has been awarded two contracts totaling \$395,000 from the Department of the Army to research the use of infrared DIAL for detection of gases of military interest.

D. Negotiations and Expressions of Interest

Negotiations with a Western European government for constructing and applying a ground-mobile visible ultraviolet DIAL are now in progress. Successful application of the ground-mobile system will lead to the construction of a versatile airborne system. A modular concept has been proposed that will incorporate infrared DIAL technology when its capabilities have been established.

SRI has prepared a research brief on the use of the DIAL technique for measuring water vapor in order to determine the radio-frequency refractive-index structure of the atmosphere. This could be very important for communication systems, since the refractive index affects the direction of propagation of electromagnetic radiation. Favorable responses have been received from a number of Department of Defense agencies and it seems likely that a funded program will ensue.

Three agencies of the U.S. intelligence community have expressed interest in the capabilities being developed at SRI for remote detection and identification of gases. Discussions are now in progress that may lead to contract support.

One important potential government user of the DIAL technique is the U.S. Environmental Protection Agency. The Research Division at Research Triangle Park, North Carolina, convened a conference on the use of lasers for remote sensing of air pollutants on 4 and 5 February 1975. Other researchers and representatives of other government agencies also attended. Both SRI and Stanford discussed results to date. It was generally agreed that the DIAL technique was the most promising active remote-sensing technique, and that it would be used in the field after further development. Subsequently, Dr. William Herget of EPA's Research Division expressed a desire to use the SRI DIAL system to measure SO_2 emissions from stacks when the system is field-qualified.

Mr. Arthur Dybdahl of the Enforcement Division of the Environmental Protection Agency, Denver, Colorado, visited SRI to discuss measurement of SO_2 using the DIAL technique. He stated that when a field-qualified DIAL system for measuring SO_2 is available, he would like to use it for enforcement purposes. The Washington office of the Enforcement Division has also expressed interest in using such a system for monitoring and abatement.

State and regional air pollution boards, including the California Air Resources Board, are also beginning to take an interest in the DIAL technique. Conversations with Dr. Peter Wagner of Maryland's Center for Environmental and Estuarine Studies indicate that the technique being

developed under this grant will be useful in smoke-plume chemistry research of interest to that state's power-plant siting program.

Industry has also displayed a keen interest in the work being done at SRI and Stanford. One prospective near-term industrial user of the research results is the electric power industry. California-based utility companies have also expressed an interest in the DIAL monitoring capabilities, and EPRI is interested in using the technique in their sulphur research project.

It is encouraging that a number of potential users in both government and industry are, or are considering, funding applications of the DIAL technique. Field use of the more versatile and more promising infrared DIAL system is imminent.

E. Applications by Others

The Environic Branch of the Air Force (formerly at Kirtland AFB, New Mexico, and now at Tyndall AFB, Florida) consulted with SRI in preparing a report, "Remote Air Pollution Monitoring and its Applicability to USAF Needs" (Captain R. A. Wallner, January 1975). The branch concluded that the DIAL technique was the most promising, and that the Air Force should consider funding development of the technique for several applications.

The Environmental Protection Agency awarded a contract to Systems Applications, Inc., to evaluate the technical and legal aspects of remote-sensing techniques for enforcement of air quality standards. SAI consulted with SRI. Their report, "Application of Remote Monitoring Techniques in Air Enforcement and Regulatory Programs" (C. B. Ludwig and M. Griggs, April 1975), concluded that the DIAL technique was the most promising approach and would be used in enforcement within several years.

NASA-Langley Research Center has a large program to develop laser remote-sensing techniques for the measurement of air pollutants, especially from aircraft and spacecraft. They have closely followed the progress at SRI and have subsequently set up similar experiments for NO_2 , SO_2 , and H_2O . They have also consulted extensively with Stanford University.

Dr. E. R. Murray of SRI has advised Molelectron, Mountain View, California; Tachisto, Inc., Needham, Massachusetts; and Lumonics Research, Ltd., Toronto, Canada, on the application of high-energy gas lasers to remote sensing and on the future direction of remote-measurement capabilities.

The EPA laboratory in Las Vegas, Nevada is developing airborne lidars using tunable dye lasers for remote measurement of air pollutants. Their work closely parallels earlier work at SRI and has benefited from information provided by SRI and developed on this grant, especially regarding fire safety.

During the past two years the research at Stanford University has led to the development of commercial instruments based on its remote monitoring system. General Photonics and Raytheon have built Nd:YAG laser-amplifier systems based on the Stanford stable-resonator source. Westinghouse is completing a parametric tuner for use by NASA-Langley Research Center. Crystal Technology now offers LiNbO_3 crystals for sale based on the crystal growth work at Stanford.

Total Technology, Inc., of San Carlos, California completed a 16-bit 10-MHz buffer memory for the Stanford remote monitoring system and made it commercially available. Also, Quanta Ray, Inc., of Mountain View has developed a Nd:YAG unstable resonator laser system based on the Stanford research. They expect to develop the high-energy LiNbO_3 tuner and associated components that use tunable sources, including a remote monitoring system, that were developed at Stanford.

Bethlehem Steel is planning to use a Stanford-designed remote-sensing system for CH_4 monitoring in coal mines. Use of this instrumentation may lead to a 20% increase in productivity and significantly improve mine safety.

IV CONCLUSIONS

During this research several lidar transmitter/receiver combinations for the measurement of specific groups of air pollutants were experimentally evaluated. These different DIAL systems operate in the ultraviolet, visible, near-infrared, mid-infrared, and far-infrared spectral regions. Specific lasers with which demonstration measurements were conducted are given below:

- Continuously-tunable dye laser for measurement of NO_2 near 440 nm
- Continuously-tunable, frequency-doubled dye laser for sensing SO_2 and O_3 near 300 nm
- Discretely-tuned deuterium fluoride laser for monitoring HCl , CH_4 , and N_2O near 3.8 μm
- Discretely-tuned CO_2 laser for measurement of H_2O , ethylene, and temperature near 10 μm
- Continuously-tunable Nd:YAG-pumped OPO for sensing SO_2 and CH_4 .

These measurements have demonstrated the range, range-resolution, and sensitivity of DIAL systems. In addition, wavelengths have been identified for measurement of many additional species using the abovementioned infrared lasers.

There is no one optimum remote-measurement system for all air pollutants, but several generalities regarding the use of each type of system have become apparent. The tunable dye and frequency-doubled dye lasers are limited to the detection of NO_2 , SO_2 , and O_3 . Range-resolved measurements of ambient concentrations with these systems are possible with current technology to a range of 2 km. To measure other important species it is necessary to use an infrared lidar. The discretely-tuned lasers have higher output energy than the continuously-tunable lasers, and thus provide longer range. However, the discretely-tuned lasers are limited to operation on a fixed set of wavelengths. This limits opportunities for selecting high-sensitivity, interference-free laser lines.

The continuously-tuned lasers therefore offer much more flexibility in selection of optimum wavelengths, but such systems operate at shorter ranges. The best lidar system for the measurement of an air pollutant will depend on:

- (1) the specific gas to be measured,
- (2) the concentration of that gas,
- (3) the concentrations of interfering species, and
- (4) the maximum range and range-resolution required for the measurement.

This research program has already led to extensive use of lidar technology by government and industry. The earlier ultraviolet/visible lidar technology initiated two generations of mobile lidars designed, constructed, and used in the field for the Electric Power Research Institute. The ultraviolet lidar technology developed under this grant has been extended under contract with the Army to remote sensing of biological aerosols. The infrared DIAL technology pioneered under this grant has led to research and development of remote sensing of military chemicals under funding by the Army. The USGS and a major oil company have supported continued development of infrared DIAL for petroleum exploration. The Air Force and NASA have supported systems studies of remote sensing capabilities from spacecraft, aircraft, and ground-mobile platforms. In addition, infrared lidar has the potential for measurement of aerosol composition using the differential scatter (DISC) technique developed at SRI. The DISC technique uses exactly the same equipment as an infrared DIAL system, thus offering the potential for simultaneous measurement of gas concentration and aerosol composition.

The applications of DIAL remote measurement systems include: enforcement of air-pollution regulations, testing of air pollution models, real-time examination of the atmospheric chemistry of air pollutants, petroleum exploration via detection of light hydrocarbons indicative of subsurface deposits, detection of pipeline leaks, detection of gases of military interest, and exploration for regions of geothermal activity.

Due to the varied applications of the research performed under this grant, this research will continue to strongly impact many different areas of the scientific applications community.

REFERENCES

- F. B. Abeles and H. E. Heggestad, J. Air Poll. Control Assoc., Vol. 23, p. 517 (1973).
- R. J. Anderson and P. R. Griffiths, J. Quant. Spectrosc. Radiat. Transfer, 17, Vol. 6, pp. 393-401 (1977).
- C. H. Bair and F. Allario, Appl. Optics, Vol. 16, pp. 97-100 (1976).
- I. J. Barton and J. F. LeMarshall, Opt. Letters, Vol. 4, pp. 78-80 (1979).
- R. A. Baumgartner and R. L. Byer, "Remote SO₂ Measurements at 4 μ m with a Continuously-Tunable Source", Opt. Letters, Vol. 2, p. 163 (1978a).
- R. A. Baumgartner and R. L. Byer, "Continuously-Tunable IR Lidar with Applications to Remote Measurement of SO₂ and CH₄", Appl. Optics, Vol. 17, p. 3555 (1978b).
- R. A. Baumgartner and R. L. Byer, "Optical Parametric Amplification", IEEE Journ. Quant. Electr., QE-15, p. 432 (1979).
- Y. Beers, Introduction to the Theory of Errors (Addison-Wesley, Mass., 1962).
- B. Bolin and C. D. Keeling, J. Geophys. Res., Vol. 68, pp. 3899-3920 (1963).
- S. E. Brosman and R. L. Byer, "Optical Parametric Oscillator Threshold and Linewidth Studies", IEEE Journ. Quant. Electr., QE-15, p. 415 (1979).
- R. L. Byer and M. Garbuny, Appl. Optics, Vol. 12, pp. 1496-1505 (1973).
- R. L. Byer and R. L. Herbst, "The Unstable Resonator Nd:YAG Laser Source", Laser Focus (July 1978).
- R. L. Byer, R. L. Herbst, and R.N. Fleming, "A Broadly Tunable IR Source", in Laser Spectroscopy, S. Haroche, J. C. Pebey-Peyroula, T. W. Hansch, and S. E. Harris, eds., pp. 207-26 (Springer-Verlag: Berlin, Germany, 1975).
- R. L. Byer and L. A. Shepp, "Two-Dimensional Remote Air Pollution Monitoring via Tomography", Opt. Letters, Vol. 4, p. 75 (1979).
- J. G. Cooney and M. Pina, Appl. Optics, Vol. 15, pp. 602-603 (1976).

- R. Gill, K. Geller, J. Farina, J. Cooney, and A. Cohen, J. Appl. Meteorol. Vol. 18, pp. 225-227 (1979).
- W. B. Grant, R. D. Hake, E. M. Liston, R. C. Robbins, and E. K. Proctor, Jr., Appl. Phys. Letters, Vol. 24, pp. 550-552 (1974).
- W. B. Grant and R. D. Hake, Jr., J. Appl. Phys. Vol. 46, pp. 3019-3023 (1975).
- K. M. Haught and J. A. Dowling, Opt. Letters, Vol. 1, p. 121 (1977).
- R. L. Herbst, H. Komine, and R. L. Byer, Optics. Commun. Vol. 21, pp. 5-7 (1977).
- J. E. Kalshoven, Jr. and C. L. Korb, NASA Technical Memorandum No. 79538 (1978).
- C. D. Keeling et al., Tellus XXVIII, pp. 538-551 (1976).
- J. B. Mason, Appl. Optics, Vol. 14, pp. 76-78 (1975).
- R. A. McClatchey et al., "AFCRL Atmospheric Absorption Line Parameter Compilation", Report No. ARCRL-TR-73-0096, L. G. Hanscom Field, Bedford, Mass. (1973).
- E. R. Murray, J. E. van der Laan, and J. G. Hawley, Appl. Optics, Vol. 15, pp. 3140-3148 (1976a).
- E. R. Murray, R. D. Hake, Jr., J. E. van der Laan, and J. G. Hawley, Appl. Phys. Letters, Vol. 28, pp. 542-543 (1976b).
- E. R. Murray and J. E. van der Laan, Appl. Optics, Vol. 17, pp. 814-817 (1978).
- R. R. Patty, G. M. Russwurm, W. A. McClenny, and D. R. Morgan, Appl. Optics, Vol. 13, p. 2850 (1974).
- S. S. Penner, J. Quant. Spectrosc. Radiat. Transfer, Vol. 18, pp. 549-553 (1977).
- A. S. Pine and P. F. Moulton, J. Mol. Spectrosc., Vol. 64, p. 15 (1977).
- K. N. Rao, Physical Chemistry, (ed. D. A. Ramsay), Butterworth, U.K., (1976).
- M. S. Shumate, R. T. Menzies, J. S. Margolis, and L. G. Rosengren, Appl. Optics, Vol. 15, pp. 2480-88 (1976).
- D. J. Spencer, G. C. Denault, and H. H. Takimoto, Appl. Optics, Vol. 13, pp. 2855-2868 (1974).
- R. G. Strauch, V. E. Derr, and R. E. Cupp, Appl. Optics, Vol. 10, p. 2665 (1971).

Appendix

PUBLICATIONS AND PRESENTATIONS UNDER THIS RESEARCH

Appendix

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- Baumgartner, R. A. and R. L. Byer, "Remote SO_2 Measurements at $4 \mu\text{m}$ With a Continuously-Tunable Source", Optics Letters, Vol. 2, No. 6 p. 163 (June 1978).
- Baumgartner, R. A. and R. L. Byer, "Continuously-Tunable IR Lidar With Applications to Remote Measurements of SO_2 and CH_4 ", Appl. Optics, Vol. 17, No. 22, p. 3555 (November 1978).
- Baumgartner, R. A. and R. L. Byer, "Optical Parametric Amplification", IEEE J. Quant. Electr., Vol. OE-15, p. 432 (1979).
- Brosnan, S. J. and R. L. Byer, "Optical Parametric Oscillator Threshold and Linewidth Studies", IEEE J. Quant. Electr., Vol. OE-15, p. 415 (1979).
- Byer, R. L., "Remote Air Pollution Measurement", reprinted from Proceedings of ICO Conference on Opt. Methods in Sci. and Indust. Measu. Tokyo, 1974, Japan J. Appl. Physics, 14, Suppl. 14-1 (1975). (See also Optical and Quantum Electronics, Vol. 7, p. 147 (1975)).
- Byer, R. L., "High-Energy Tunable IR Source and Applications", Proc. SPIE, Vol. 95, Modern Utilization of Infrared Technology II, p. 92 (1976).
- Byer, R. L., "Three-Dimensional Monitoring: Differential Absorption Laser Radar", published in Electric Power Research Institute Journal, p. 45 (July/August 1976).
- Byer, R. L., "High-Energy Tunable Source and Applications", SPIE Conference, San Diego, California (August 1976).
- Byer, R. L., "Remote Detection of Air Pollutants", Conference on Lasers and Applications, Washington, D.C. (May 1977).
- Byer, R. L., "Remote Monitoring of Atmospheric Pollutants with a Continuously-Tunable Source", International Laser/Radar Conference, Philadelphia, Pennsylvania (June 1977).
- Byer, R. L., "Remote Air Pollution Monitoring", International Air Pollution Conference, Rjukan, Norway (1978).

- Byer, R. L., "Lasers for Air Pollution Monitoring", Sonoma State College, Sonoma, California (October 1978).
- Byer, R. L., "Remote Air Pollution Measurements with Lasers", Los Alamos Scientific Laboratory, Los Alamos, New Mexico (December 1978).
- Byer, R. L., "Remote Infrared Lidar Measurements of SO_2 and CH_4 --Recent and Future Capability", presented at the A.G.U. meeting, Washington, D.C., May 28-June 1, 1979.
- Byer, R. L., "High-Energy Tunable Lasers with Applications to Remote Air Pollution Measurements", presented at the 9th International Laser Radar Conference, Munich, Germany, July 2-5, 1979.
- Byer, R. L., "Infrared Lidar Absorption Measurements in the Atmosphere", presented at the 6th F.A.C.S.S. Conference, Philadelphia, Pennsylvania September 16-20, 1979.
- Byer, R. L. and M. Endemann, "Remote Probing of Atmospheric CH_4 Over Long-time Scales and High Accuracy Using a Widely-Tunable IR Source", presented at the 9th International Laser Radar Conference, Munich, Germany, July 2-5, 1979.
- Byer, R. L. and E. R. Murray, "Remote Monitoring with Laser Sources" in Air Pollution Analysis, R. Perry, ed. (Chapman and Hall, Ltd. London, 1977).
- Grant, W. B., R. D. Hake, Jr., E. M. Liston, R. C. Robbins, and E. K. Proctor, Jr., "Calibrated Remote Measurement of NO_2 Using the Differential-Absorption Backscatter Technique", Appl. Phys. Letters, Vol. 24, No. 11, pp. 550-552 (June 1974).
- Grant, W. B. and R. D. Hake, Jr., "Calibrated Remote Measurements of SO_2 and O_3 Using Atmospheric Backscatter", J. Appl. Phys., Vol. 46, No. 7, p. 3019 (July 1975).
- Murray, E. R., "Remote Measurement of Gases Using Discretely-Tunable Infrared Lasers" (invited paper), Proceedings of the Society of Photo-Optical Instrumentation Engineers, Vol. 95 (September 1976).
- Murray, E. R., "Remote Measurement of Gases Using Discretely-Tuned Infrared Lasers", presented at the Applied Physics Colloquium, Stanford University, Stanford, California (November 1976).
- Murray, E. R., "Air Pollution Measurements Using Differential-Absorption Lidar", presented at the Seminar on Air Pollution Monitoring, Dutch Institute of Public Health, Bilthoven, Holland (December 1976).
- Murray, E. R., "Remote Measurement of Gases Using Discretely-Tunable Infrared Lasers", Opt. Engrg., Vol. 16, No. 3, p. 284 (May 1977).

- Murray, E. R., R. D. Hake, Jr., J. E. van der Laan, and J. G. Hawley, "Atmospheric Water Vapor Measurements with an Infrared (10- μ m) Differential-Absorption Lidar System", presented at the 7th International Laser Radar Conference, Menlo Park, California, (September 1975).
- Murray, E. R., R. D. Hake, Jr., J. E. van der Laan, and J. G. Hawley, "Atmospheric Water Vapor Measurements with an Infrared (10- μ m) Differential-Absorption Lidar System", Appl. Phys. Letters, Vol. 28, No. 9, p. 542 (May 1976).
- Murray, E. R., J. E. van der Laan, J. G. Hawley, R. D. Hake, Jr., M. F. Williams, "Remote Sensing of Gases Using Single-Ended Differential-Absorption Lidar Systems", Proceedings of the Workshop on Remote Sensing of the Marine Boundary Layer, Vail, Colorado (August 1976).
- Murray, E. R., J. E. van der Laan, J. G. Hawley, R. D. Hake, Jr., and M. F. Williams, "DIAL Systems for Monostatic Sensing of Atmospheric Gases", Proceedings of the 8th Materials Research Symposium, National Bureau of Standards, Gaithersburg, Maryland (September 1976).
- Murray, E. R., J. E. van der Laan, and J. G. Hawley, "Remote Measurement of HCl, CH₄, and N₂O Using a Single-Ended Chemical Laser Lidar System", Appl. Optics, Vol. 15, No. 12, p. 3140 (December 1976).
- Murray, E. R., M. F. Williams, and J. E. van der Laan, "Single-Ended Measurement of Infrared Extinction Using Lidar", Appl. Optics, Vol. 17, No. 2, p. 296 (January 1978).
- Murray, E. R. and J. E. van der Laan, "Remote Measurement of Ethylene Using a CO₂ Differential-Absorption Lidar", Appl. Optics, Vol. 17, No. 5, p. 814 (March 1978).